MINERALOGICAL MAGAZINE

JOURNAL OF THE MINERALOGICAL SOCIETY

Vol. 32 September 1959 No. 246

Leonard James Spencer.

By W. Campbell Smith.

ON rare occasions the Mineralogical Society has published obituary notices, with a full-page portrait, of its more distinguished officers on their decease. Such notices were written for Henry Clifton Sorby, first President of the Society, Nevil Story-Maskelyne, Sir Lazarus Fletcher, Sir Henry Miers, and Frederick Noel Ashcroft.

These were very special tributes to men who had devoted themselves to the work of the Society and to the study of our science. No one, surely, has so highly deserved such a tribute as Leonard James Spencer, editor of the 'Mineralogical Magazine' from 1901 to 1955 and of 'Mineralogical Abstracts' from its beginning in 1920 to 1955. He was President of the Society from 1936 to 1939, and Foreign Secretary from 1949 until his death on 14 April 1959 at the age of eighty-eight.

Two other tributes to Dr. Spencer have appeared recently in the 'Mineralogical Magazine'. One is an account of his work at the British Museum (Natural History), published in the Jubilee Volume issued in 1950 to mark the fiftieth year of his editorship (vol. 29, p. 256); the other, 'An Appreciation' written by Dr. James Phemister, appears in the first number of the Magazine after Spencer's retirement in 1955 (Min. Mag., vol. 31, p. 1). It is difficult to add anything to these accounts unless one attempts to write a little more intimately of his work at home.

Spencer was born at Worcester on 7th July 1870, the eldest son of James Spencer, who was for many years headmaster of the School department of Bradford Technical College. He and his brothers were brought up to work seriously and to spend little. His father presented Leonard James with a geological hammer at the age of seven, and at once the boy seems to have started to make a collection of minerals, rocks, and fossils. This collection by 1892 contained some 3000 labelled and catalogued specimens, and was offered to, and gratefully accepted by, Bradford Technical College for the use of their students.

By this time Spencer was in his third year at Sidney Sussex College, Cambridge. When only sixteen he obtained a Royal Exhibition, which took him to the Royal College of Science for Ireland in Dublin, where he gained first-class honours in chemistry in 1889. His geology professor in Dublin was J. P. O'Reilly, who was succeeded by Grenville Cole in the year after Spencer left.

Spencer went on to Cambridge with a scholarship to Sidney Sussex and was there four years, taking a first in both parts of the Tripos and winning the Harkness Scholarship for Geology in 1893. He was the first Sidney Sussex man to win this high award, but he set a fashion in his College, for no fewer than six Sidney men have won it since.

Funds were very short for Spencer at College, and he lived quietly and kept very much to himself. Already, at Cambridge, he was abstracting papers for the Patents Office, and so set out, perhaps from necessity, on a work that was later to become a labour of love.

It was fortunate for British Mineralogy that a vacancy in the Department of Mineralogy at the British Museum was being advertised just at the end of Spencer's fourth year at Cambridge. It was fortunate also that Spencer defeated the other three candidates in the examination. The recollection that one of these was the late Sir William J. Pope was a source of amused satisfaction to Spencer in later years.

Before taking up his post at the Museum Spencer went, at Fletcher's suggestion but at his own expense, to Munich to study crystallography under Paul Groth. He arrived in Munich on 30 September 1893; lectures did not begin until November, but he spent the intervening month in the laboratories and in learning German from a teacher, an hour and a half each day. When term began he attended Groth's lectures, and also Weinschenk's on petrology. Leaving Munich on 18 December he travelled back via Heidelberg, arriving home for Christmas Day, and reported for duty at the Museum on New Year's Day, 1894.

His work at the Museum has been fully described, and Dr. Phemister has told how he began at once contributing abstracts to the 'Mineralogical Magazine', and voluntarily prepared an index to the first ten volumes. Gradually he did more and more for the Society, but this did not interfere with his Museum work. He worked long hours at home. He rarely took a holiday, and when he did it would usually be for a collecting trip to some English mines.

Spencer made three long trips abroad primarily for the purpose of collecting specimens and when possible of visiting museums and private collections. One of these trips was to Canada and the U.S.A. in 1924 for

the British Association Toronto meeting; the second to South Africa and South-West Africa in 1929 for the British Association and International Geological Congress; and the third, a real adventure, was to the Libyan desert. On this occasion he accompanied the expedition organized by P. A. Clayton and the late O. H. Little to investigate the mysterious silica-glass *in situ*. Spencer was already sixty-four, but he had no qualms about the desert journey, and the only stipulation he made was that he should have a tent to himself at night.

In 1899 Spencer married Edith Mary, daughter of Islip J. Close of Mortimer, Berkshire. Mrs. Spencer was a very gifted woman, able in the early years of their married life to help with the translation of Max Bauer's great quarto 'Edelsteinkunde'. They had a son and two daughters, and it must have been with some slight surprise, and certainly with pride, that Spencer saw his son creating a flourishing engineering business and his two daughters, both very talented, rising to the front ranks of their profession, each in her own special line, one in ballet and the other in orchestral music. Spencer was very fond of little children and he took great pleasure in his numerous grandchildren and his three great-grandchildren, and he never forgot a birthday.

Spencer had few hobbies other than collecting minerals and writing abstracts. He was, however, a keen gardener and spared a little time for this, cultivating the small garden at his house in London and brightening the view in front for the passer-by with gay flowers, or, on occasions and by way of a joke, with great gourds. For some years he had a cottage at Chobham where the family went for week-ends, and Spencer worked energetically cultivating flowers and vegetables in the narrow quarter-mile-long garden. The products of the garden were conveyed to London in heavy sacks, Spencer making the first part of the journey to Woking station with a bag on each handlebar of his strongly-built bicycle. For a time also he possessed a motor launch, which was moored near his house at Albert Bridge. This he had bought primarily for the amusement of his son, but it afforded Spencer plenty of occupation too, and on occasions, when the engine gave out, exercise as well.

During the two wars he continued doggedly with his work at home and at the Museum, and in 1914 to 1918 he added to his other duties those of a Special Constable in his home district. In the Second World War, refusing to leave the house, he slept at nights under the table in his dining room. It was only under compulsion that he consented to move for a few nights to a neighbouring underground shelter while an unexploded bomb was removed from the garden of his house. He

suffered much anxiety at that time for the safety of his indexes and library.

As the family grew up, married, and left home, the 'Mineralogical Magazine' and 'Abstracts' and the vast collection of reprints encroached on more and more rooms in the four-storey house by Albert Bridge. The triennial indexes to the 'Abstracts' were compiled as the abstracts were printed off, countless slips being housed in home-made but quite efficient cabinets, made partly from wood salvaged off the beach by Albert Bridge. When the moment came to sort the slips and prepare them for the printer, more of the house and some of the family, if available, would become involved in the work. A daughter might be prevailed upon to sort the slips alphabetically and then, when duplicates had been eliminated, the edited slips were pasted on strips cut from issues of 'The Times' and laid out on the stairs to dry. All this, like so much of Spencer's editing, was planned to save the Society expense. It may have appeared make-shift, but it was efficiently organized and it was not time-wasting.

Most of those who had papers through his hands knew him as a most meticulous and persistent editor. He had a remarkable capacity for attention to every detail of a paper, and he had a sufficiently good knowledge of many languages and a good enough memory to enable him to detect every flaw in the spelling or printing of titles or text. At meetings he usually spoke on some point of detail of nomenclature or locality, and he seldom did himself justice when reading a paper or speaking in public. No one who worked with him could fail to appreciate the depth of his knowledge, his limitless industry and power of concentration, and, beneath a somewhat brusque manner, a saving sense of humour and a truly kind heart.

X-ray studies of synthetic diamonds.

By Kathleen Lonsdale, D.B.E., D.Sc., F.R.S.,

H. Judith Milledge (née Grenville-Wells), M.Sc., Ph.D.,

and

Eric Nave.

University College, London, W.C. 1.

(With Plates IX and X.)

[Read 2 April 1959.]

Summary. Studies are described of various specimens of diamonds made by the General Electric Company of New York, and by Allmänna Svenska Elektriska Aktiebolaget, Västeras, Sweden. The G.E. synthetic diamonds always contain single crystal inclusions of nickel or of a Ni-rich face-centred cubic compound. These are strictly parallel to the diamond surrounding them. If the nickel inclusions are twinned on (111), the diamond is similarly twinned; and this is usually the case. It is suggested that epitaxial growth is part of the mechanism of the graphite-to-diamond transformation in the G.E. technique. Other powdered or single-crystal inclusions are also found; some of those correspond to compounds that occur in meteoritic carbon. There is no parallel growth in such cases. The Swedish synthetic diamonds do not contain nickel, but they are generally less well-crystallized than the G.E. specimens. Both, however, give good diffraction spots indicating a well-ordered structure with (except very rarely) no trace of graphite present.

THIS work may be grouped under three headings: studies of diamonds made by the General Electric Company of New York, U.S.A. (hereafter referred to as G.E. diamonds); studies of diamonds made by the Allmänna Svenska Elektriska Aktiebolaget, Västeras, Sweden (ASEA diamonds); and a further study of a Hannay diamond.

The work is not as complete as we would have liked, but it seems possible that further investigation would be a waste of time, since presumably studies at present classified as secret will eventually be released for publication.

G.E. Diamonds.

These diamonds have been obtained from three sources: those that one of us (H. J. M.) was kindly allowed to select during a lecture visit to the G.E. Research Laboratories, Schenectady, New York; those sent from the G.E. Research Laboratories to various research centres in

Great Britain, including our own; and those sold in the U.S.A. and given to us by Mr. P. Neven of the Impregnated Diamond Products Ltd., Gloucester.

The diamonds are formed at pressures up to 100 000 atm. and temperatures up to 3000° K. The G.E. Research Information Services released a press statement 'Man-Made Diamond' in February–March 1955 containing articles by F. P. Bundy, H. T. Hall, A. L. Marshall, A. J. Nerad, H. M. Strong, and a résumé of the scientific literature of diamond making. A further article (Bundy *et al.*, 1955) made it clear that in addition to high pressures and temperatures, maintained over a controllable period of time in a 1000-ton press, certain 'chemical conditions' were also involved in the process of manufacture. At the time of writing, further details are still classified. In October 1957 the G.E. announced that up to that date they had made 100 000 carats of diamond powder and expected to make over 1 000 000 in 1958, the current price then being \$3.48 per carat, as against about \$2.80 per carat for corresponding natural diamond grit. The 24 % higher price for synthetic diamond seems not to be a deterrent to their sale, however, because the grinding efficiency of the synthetic diamond is said to exceed that of natural diamond by 35 %. We believe that this may be because the natural diamond grit contains cleavage splinters (C. B. Slawson, 1957) whereas the man-made diamonds, though largely irregular fragments, are comparatively uniform and are mostly twinned. The largest stone announced by G.E. measured about 1·2 mm. in its longest dimension. None of those available to us measured more than about $\frac{1}{3}$ mm. Very few were colourless; they were mostly a dirty white, grey, or brown, and badly shaped, but there was occasionally a green or yellow specimen tending towards an octahedral habit, and sometimes fairly well-formed black specimens consisting of parallel cubes were found. We are informed that reproducible variations of colour and habit can now be obtained and that, in particular, batches consisting almost entirely of cubes with good faces can be made to order. It is said to be difficult, though it is possible, to deposit diamond on a diamond seed. Gem-quality crystals have not been reported.

X-ray examination.

X-ray studies have included random rotation photographs, followed by Laue, rotation, and Weissenberg photographs of orientated single and twinned specimens from various sources, using Cu-$K\alpha$ and β, Mo-$K\alpha$, Cr-$K\alpha$, and Ni-$K\alpha$ radiation. Powder and single (including

twinned) crystal photographs of natural diamond have been taken for comparison purposes.

Random rotation photographs taken with unfiltered Cu-K radiation showed the normal diamond pattern but, in addition, every G.E. diamond examined showed reflections in the 200 position that would not be expected for diamond, together with satellite spots on the high-angle side of all the diamond reflections (pls. IX and X). These satellite reflections were found to form the pattern of a face-centred cubic structure having a lattice constant of 3·539 Å. as compared with 3·567 Å. for the diamond (H. J. Grenville-Wells and K. Lonsdale, 1958*a*). The latter spacing is identical with that for natural diamond and no variation has been observed greater than the limit of experimental error, $\pm 0{\cdot}1$ %. The density of G.E. diamond as measured by flotation of individual crystals in diluted Clerici solution is 3·517 g./c.c. at room temperatures, which is very close to the natural diamond calculated value of 3·514. There are, however, some G.E. diamonds which float in methylene iodide (density less than 3·3 g./c.c.) and yet give an X-ray pattern of normal lattice constant. The cause of this very low density has not been found but may simply be cracks; if it were due to vacant sites randomly arranged in the structure, these would be expected to show up as a strong background intensity, which is not observed.

Orientated rotation photographs confirmed the parallelism of the diamond and satellite structures. All such photographs showed the satellite structure, although with varying relative intensity; there were never any satellite spots in positions other than those given by a structure absolutely parallel to the diamond. These satellite spots are also, in general, as sharp as those of the diamond except that there is a small $\Delta\theta$-broadening at high angles, indicating that the satellite inclusions are under uniform strain. Occasionally the satellite spots are a little more drawn-out along constant-θ arcs than those of the diamond but if the diamond spots show the presence of variable orientation, then the satellite spots invariably do so also. A particularly interesting feature is shown in pl. IX, fig. 4, where the 222 'forbidden' diamond reflection appears accompanied by an even stronger 222 satellite spot.

Weissenberg photographs further confirmed the parallelism of the diamond and satellite structures and provided some intensity data. Using Mo-K radiation the satellite structure is not apparent except for the 200, 420, and other forbidden reflections, because the lattice constant is such that its satellite reflections coincide with those of the $K\alpha_2$ components of the diamond spots.

There are *alternative explanations of the satellite spots.* The first is that they are due to a structure that is geometrically cubic of side 3·54 Å. but of rhombohedral symmetry only, and is intermediate between

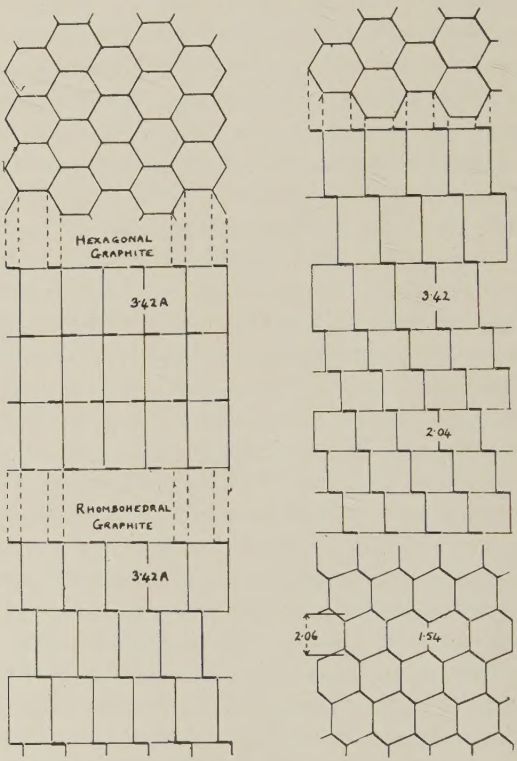


FIG. 1. Diagram to show the relationship between hexagonal and rhombohedral graphite and the possible transition from rhombohedral graphite through an intermediate structure to diamond (Grenville-Wells and Lonsdale, 1958*a*).

diamond and rhombohedral graphite (fig. 1), having carbon atoms in positions

$$000,\ 0\tfrac{1}{2}\tfrac{1}{2},\ \tfrac{1}{2}0\tfrac{1}{2},\ \tfrac{1}{2}\tfrac{1}{2}0 \pm \left|\tfrac{1}{6},\ \tfrac{1}{6},\ \tfrac{1}{6}\right|$$

and therefore closely related to the diamond structure

$$000,\ 0\tfrac{1}{2}\tfrac{1}{2},\ \tfrac{1}{2}0\tfrac{1}{2},\ \tfrac{1}{2}\tfrac{1}{2}0 \pm \left|\tfrac{1}{8},\ \tfrac{1}{8},\ \tfrac{1}{8}\right|$$

being derived from it by a straightening of the carbon atom layers parallel to one set of (111) planes only (cf. N. Nath, 1935, H. Lipson, and A. R. Stokes, 1942). The relationship to the rhombohedral graphite

structure would be that the [111] axis would have been compressed to a length of 6·11 Å. instead of the normal 10·26 Å., the graphite layers thus being 2·04 Å. apart instead of 3·42 Å. This 'intermediate structure' would have structure factors:

$$F(hkl) = 8f_c \cos \pi (h+k+l)/3$$
$$= 8f_c \text{ if } h+k+l = 3n, \quad \text{but } 4f_c \text{ if } h+k+l = 3n \pm 1.$$

Thus the intensity of the 111, $\bar{2}20$, $31\bar{1}$, &c. reflections should be four times as great as that of $\bar{1}11$, 200, 220, 311, $\bar{3}11$, &c. In other words, the 200 reflection should be relatively weak compared with other satellite spots. In fact it is relatively strong (table I); but this does not rule out the possibility that such a structure may contribute to the satellite intensities. It cannot wholly explain them.

The second possibility is that the satellite structure is that of Ni or of some Ni-rich compound. β-Ni has a face-centred cubic structure of lattice constant 3·523 Å. at room temperatures and pressures; its value at the high temperatures and pressures used is not known to us. Ni is not normally an impurity in natural diamonds. Spectrographic analysis shows, however, that it is present in all the specimens of G.E. diamonds that we were able to send for examination and present in considerable quantity in the G.E. diamonds sold for industrial purposes, even after cleaning treatment designed to remove extraneous impurities (see Appendix). That the satellite spots are due to β-Ni alone is not possible because of the strength of the 200 reflection (table I), but that they are due to a Ni-rich compound can easily be proved by taking X-ray photographs with various incident radiations. The linear absorption coefficients of C and Ni respectively for the incident radiations used are as follows:

	Mo-$K\alpha$	Cu-$K\beta$	Cu-$K\alpha$	Ni-$K\alpha$	Cr-$K\alpha$
C ...	2	12	16	23	53
Ni ...	430	2640	430	530	1220

Those of carbon containing a small percentage of nickel and of nickel surrounded by a carbon matrix will differ slightly but not appreciably from these values (H. J. Grenville-Wells and K. Lonsdale, 1958*b*). It was found that the 200 satellite spots, which are the most easily observed, are relatively strong (compared with the diamond spots) for Mo-$K\alpha$ but weak for Cr-$K\alpha$. The most convincing evidence, however, is obtained from an X-ray tube having a copper target contaminated with nickel. The amount of contamination is such that the Ni-$K\alpha$ and Cu-$K\beta$ spots, which are at approximately equal distances on each side

of the Cu-$K\alpha$ reflection, are of comparable intensities. The relative intensities for various diamond reflections for the three wave-lengths 1·66 Å. (Ni-$K\alpha$), 1·54 Å. (Cu-$K\alpha$), 1·39 Å. (Cu-$K\beta$) give mean values for the proportions in the incident radiation as follows:

$$I_0(\text{Ni-}K\alpha) : I_0(\text{Cu-}K\alpha) : I_0(\text{Cu-}K\beta) = 9 : 100 : 16.$$

The Ni-$K\beta$ spots were, in general, too weak for observation, but were in any case not needed; W-L spots were also present, due to tungsten contamination, but were not used.

The observed values of the satellite spot intensities, normalized to the Cu-$K\alpha$ 200 intensity, are given in Table I. As will be seen, the Cu-$K\beta$

TABLE I. Observed ratios of satellite spot intensities, taking the Cu-$K\alpha$ 200 spot as 100, compared with the ratio of incident intensities, I_0.

	111	200	022	311	222	400	133	420	422	I_0
Ni-$K\alpha$	5·0	9·4	3·0	2·9	1·6	0·4	..	..	..	9
Cu-$K\alpha$	53·8	100	36·0	25·4	15·5	9·6	8·8	16·0	..	100
Cu-$K\beta$	1·4	5·4	0·7	1·0	0·6	0·4	..	..	..	16

intensity is markedly weakened (filtered out) relative to the Ni-$K\alpha$. This definitely proves the presence of Ni in the subsidiary structure, and the actual intensities, together with the sharpness of the satellite spots, can be used to give an approximate idea of the size of the inclusions, which appear to be of the order of 5.10^{-4} cm. linear dimensions (H. J. Grenville-Wells and K. Lonsdale, 1958*b*). The G.E. diamonds, when examined at high magnification, are seen to contain so much impurity and other irregularities that it is not possible to verify the presence of these nickel-rich inclusions visually.

We have previously suggested that the subsidiary structure may consist of the cubic form of $Ni_xC (x > 4)$ reported by R. Bernier (1951).[1] If this is so, the intensities observed would indicate some randomness of both Ni and C in a structure having a partial rock-salt type character. Attempts to repeat the synthesis of cubic Ni_xC for intensity-comparison purposes have not been successful but in any case the inclusions in the G.E. diamonds may have been formed under conditions of high pressure and temperature not reproducible in an ordinary laboratory.

Some surprise had been felt that so small an amount of Ni as 0·2 % should give easily visible diffraction spots. A mixture of diamond and Ni powder was therefore made in as near this proportion as was possible. The X-ray pattern of this mixture did indeed show the Ni lines quite

[1] Distances given in this paper as Å. are really kX; to correct to Å. multiply by 1·00202.

clearly (pl. IX, fig. 5). The diffraction effect of the impurity would be expected to be even clearer where it is concentrated into a few single crystals.

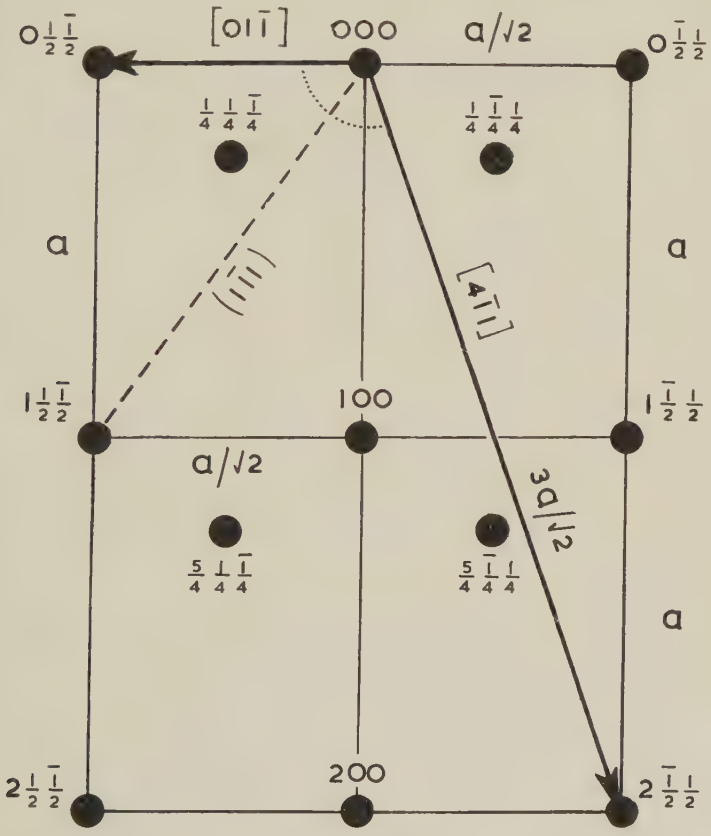


FIG. 2. The axis [01$\bar{1}$], identity period $a/\sqrt{2}$, twins across (1$\bar{1}$1) into [4$\bar{1}$1], identity period $3a/\sqrt{2}$. The positions of carbon atoms actually lying in the (011) plane through the origin are shown. Compare table II and pl. IX, figs. 1–4 for the reflection spots found for a rotation about [01$\bar{1}$] and [4$\bar{1}$1] of a twinned crystal.

The G.E. diamonds are almost invariably twinned on (III), although the components may be of very unequal magnitudes. Whatever the degree of twinning, it seems to be paralleled by the Ni-rich inclusions which are also, in that case, twinned on (III). Pl. IX, figs. 1–4 show this very clearly. The diamond was set to rotate about a [01$\bar{1}$] edge. The layer lines on the photograph so obtained correspond to an identity

distance of $3a/\sqrt{2}$ instead of to $a/\sqrt{2}$, ($a = 3{\cdot}567$ Å.). The first and second layer lines, which are due to one member of the twin only, may be of weak intensity compared with the zero and third layer lines, for both diamond and Ni structures. Fig. 2 shows that the $[01\bar{1}]$ reflects across $(1\bar{1}1)$ to coincide with $[4\bar{1}1]$. The reflections occurring on the various layer lines will be as shown in table II (those due to the twin

TABLE II. Indices of the spots observable on an X-ray diffraction photograph of a twinned crystal rotating about $[01\bar{1}]$ of one member of the twin; this is $[4\bar{1}1]$ of the other. Indices for the second member underlined.

$k-l=2$	$11\bar{1}$	020	220	131	$31\bar{1}$	—	—	331	$3\bar{1}\bar{3}$
	$\underline{11\bar{1}}$	$\underline{00\bar{2}}$	$\underline{20\bar{2}}$	$\underline{\bar{1}31}$	$\underline{3\bar{1}\bar{1}}$			$\underline{\bar{3}31}$	$\underline{\bar{3}1\bar{3}}$
	$1\bar{1}1$	—	220		$1\bar{3}\bar{1}$	—	—	—	
$4h-k+l=6$			$\underline{20\bar{2}}$						
$4h-k+l=4$	111	—	$0\bar{2}\bar{2}$		—	$22\bar{2}$	004	133	
	$\underline{1\bar{1}\bar{1}}$						$\underline{0\bar{4}0}$		
$4h-k+l=2$	$11\bar{1}$	002	—		131	—	—	—	
		$\underline{0\bar{2}0}$			$\underline{1\bar{1}\bar{3}}$				

$k-l=0$	111	$\bar{1}11$	200	022	311	$\bar{3}11$	222	$\bar{2}22$	400	133	$\bar{1}33$
	$\underline{1\bar{1}\bar{1}}$	$\underline{11\bar{1}}$	$\underline{\bar{2}00}$	$\underline{0\bar{2}2}$	$\underline{3\bar{1}1}$	$\underline{3\bar{1}1}$	$\underline{2\bar{2}2}$	$\underline{2\bar{2}2}$	$\underline{\bar{4}00}$	$\underline{\bar{1}33}$	$\underline{1\bar{3}3}$
	—	—	—	022		$13\bar{1}$		—	—	—	
$4h-k+l=0$				$\underline{0\bar{2}2}$		$\underline{\bar{1}31}$					

rotating about $[4\bar{1}1]$ are underlined). A pattern of this kind (with 200 absent) can of course be obtained from a natural diamond twin. The point to be emphasized here is the exact parallelism in the intensity distribution as well as in spot positions for the G.E. diamond and Ni structures. We have here a twinned inclusion within a twinned diamond, the twin law and the approximate proportions of the individuals being the same in each case. It is a significant fact that no trace of *free* Ni has been found by X-rays and that the G.E. diamonds are strongly attracted by an electromagnet (B. W. Anderson, 1958). They do not appear in general to give X-ray fluorescence in a Cu-$K\alpha$ incident beam and this confirms the comparative absence of iron shown by the spectrographic analysis (but see below, p. 196). The general fluorescence of a random sample in ultra-violet light is surprisingly normal.

The graphite→diamond transformation would seem in the case of the G.E. process to depend upon the presence of this Ni-rich parallel inclusion. It is impossible to avoid the conclusion that the diamond has grown epitaxially upon the nickel. Fig. 3 shows that the structures of

the octahedral planes of nickel and of diamond are closely similar in dimensions. The layer plane of graphite is similarly almost identical with the projection of the puckered hexagons of diamond {111}. A possible mechanism of transformation is shown in fig. 4. According to H. T. Hall (1958) the sequence of events in the high-pressure, high-temperature synthesis in the laboratory is as follows: the high pressure

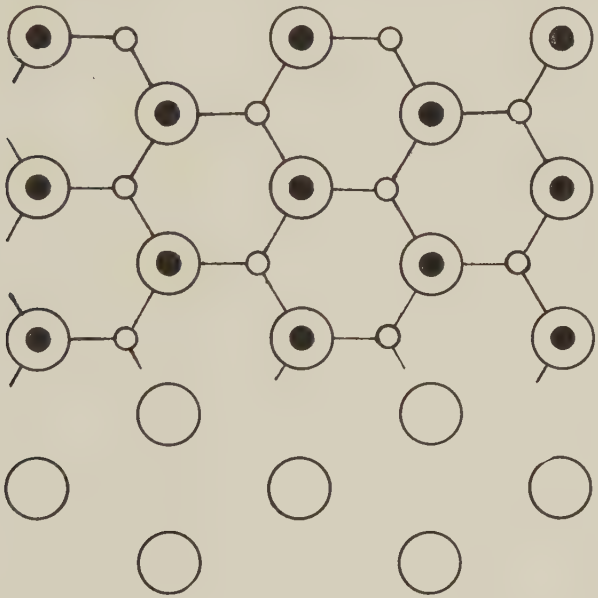


FIG. 3. Correspondence between (111) planes of diamond (or rhombohedral graphite) and of Ni or Ni_xC. Large circles: Ni atoms. Small circles: C atoms; one level for graphite, two levels for diamond.

is first applied to the starting material; then, maintaining the high pressure, the material is subjected to a temperature sufficiently high to obtain a reasonable rate of transition or reaction (the pressure and temperature together being such that the end-product, which would be only metastable at NTP, is stable); next, the temperature is decreased to room temperature; and finally the pressure is decreased to atmospheric.

In another publication H. T. Hall (1956) states that purely thermodynamic considerations 'indicate that diamond can not be formed directly from graphite. Experiment confirms this conclusion at pressures to 100,000 atmospheres and temperatures to approximately 3000° C.'

The sequence of events in the graphite→diamond transformation would seem to be: First, the high pressure brings the graphite layers much closer together, at the same time causing sideways shifts in favour of the rhombohedral form in which there is a repeat after three layers

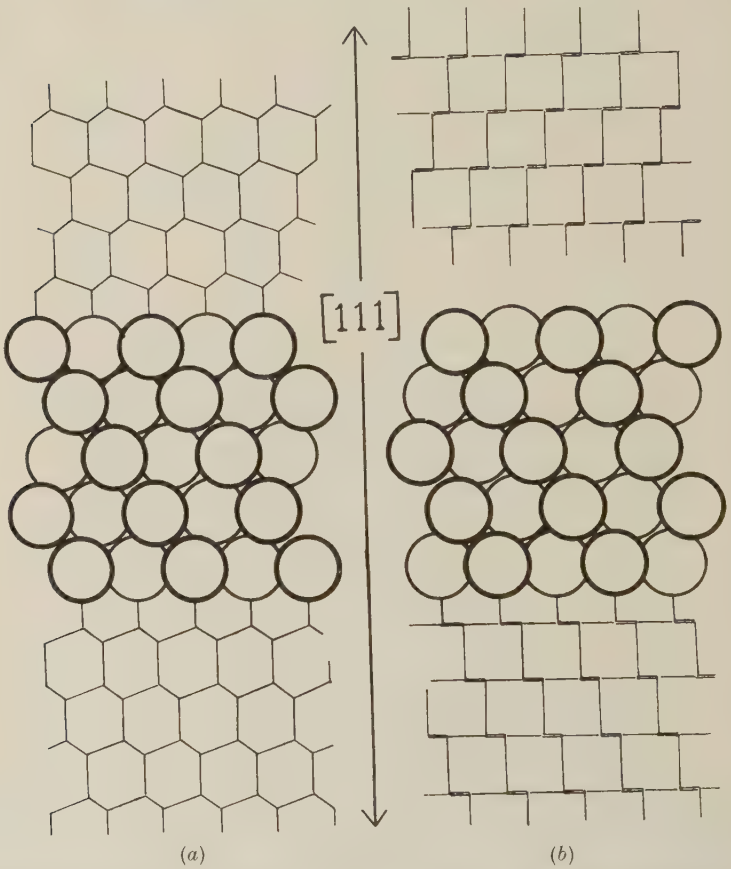


FIG. 4*a* (*left*): Growth of diamond twin on Ni base. 4*b* (*right*): Graphite under pressure, in relation to the Ni base.

instead of after two as in the hexagonal form (see fig. 1; G. E. Bacon (1950) has proved that the hexagonal→rhombohedral graphite transformation may be brought about by mechanical working); when the graphite layers are pressed close to the octahedral face of a nickel grain, alternate carbon atoms attach themselves firmly to nickel atoms (the fact that nickel carbides can be formed shows that such bonds are

possible); or it may be that the attachment is to a surface carbon layer in a cubic form of Ni_xC, if this exists prior to the experiment. The imposition in the next stage of a high temperature, the high pressure being maintained, would have two possible effects: some carbon atoms might diffuse through the nickel grains to form Ni_xC, having a lattice constant slightly greater than that of pure cubic Ni; and the graphite layers would be violently agitated although they could not separate. Since each alternate carbon atom has attached itself to the Ni atoms in an octahedral face of the included Ni grains, only the unattached, intermediate carbon atoms would be free to move permanently out of the plane of the attached graphite layers. This could initiate the formation of diamond, the plane layers becoming puckered and the highly-compressed van der Waals bonds between layers becoming normal C–C diamond bonds. A lowering of temperature, pressure still being maintained, would stabilize the diamond structure at room temperature around the nickel inclusions, the reversion to graphite being prevented by the continued compression. The high pressure could then be safely reduced to atmospheric.

One important question that remains is how the parallelism of graphite (0001) layers and of the octahedral planes of the Ni-rich inclusions is achieved in the first place, and how such relatively large quantities of Ni can be absorbed and not show even greater filtering of the Cu-$K\beta$ component of the incident radiation. If the Ni inclusions were even approximately spherical and of radius 5.10^{-4} cm. the overall intensity of the satellite spots and the relatively high concentration given by the spectrographic analysis would both indicate that the number of parallel crystalline inclusions present in a diamond of radius 0·3 mm. must be of the order of twenty. Admittedly the presence of twinning makes the calculation difficult because the proportion and distribution of diamond contributing to the measured spot intensities is not precisely known, but it cannot be far wrong. Possibly the existence of the Widmanstätten patterns in nickel-iron meteorites gives a clue. In a strong magnetic field graphite will orientate itself with the layers of carbon atoms parallel to the lines of force. Nickel filings, on the other hand, or beads of molten Ni recrystallizing, would orientate themselves along the lines of force. Since it is necessary to begin with single crystal material it may be that the necessary parallelism is achieved magnetically, perhaps with some initial annealing of the nickel or nickel-rich crystallites in a magnetic field.

The suggestion that the nickel may be growing in cracks in the

diamond structure does not explain the invariable parallelism, nor the need for nickel at all; and it also leaves the process of transformation unexplained. Since Ni is not found in natural diamonds, however, it cannot be essential to the process. Habit and colour will also depend upon the presence of traces of other impurities.

Inclusions in powdered form gave powder lines on a number of the specimens examined. None of these was identified. One of the G.E. diamonds sent to Birkbeck College, London, had exceptionally streaky Ni-satellite spots (indicating more disorientation than was normal for the inclusions). This also showed continuous powder lines corresponding to spacings 8·0, 3·68 Å., &c. Similar lines were found for a green octahedron from the University College batch, which was also the only G.E. diamond showing any trace of a diffraction line due to graphite. Similar lines had previously been observed from a natural diamond.

One University College specimen, consisting of a group of fairly well-formed parallel black cubes (not unlike cliftonite in appearance) agreed with a greyish-black granular fragment in the batch returned from spectrographic analysis 1 (see Appendix) in showing a strong line at 3·08 Å. and many others corresponding to smaller spacings. This black specimen showed a really remarkable parallelism between diamond and satellite spots (pl. X, fig. 1), the latter being very sharp indeed except at high angles. (Cliftonite has been studied by Grenville-Wells, 1952.)

One twin consisting of two interpenetrating cubes with excellent cube faces showed no inclusion except the usual Ni, but did give high background scattering with Cu-K radiation, which suggests the presence of iron. This is also common in Congo diamonds, the only natural source from which cube faces (usually imperfect) are frequently found.

Small foreign crystals were also found, associated with the diamonds in specimens supplied or purchased. None of these was identified. For example, among the G.E. diamonds sent to Birkbeck College was a somewhat flattened, badly-shaped octahedron with a black matt surface, which gave no diamond or Ni spots, but gave a powder pattern leading to interplanar spacings:

2·95 Å., *w*; 2·53, *ms*; 2·07, *w*; 1·72, *vvw*; 1·475, *ms*;
1·275, *w*; 1·085, *mw*; 1·00, *w*; 0·88, 0·855, &c., all *vvw*.

Some of these lines were also found from a red crystal, also not diamond, in a sample of G.E. diamonds sent to the Royal Institution, London. This red crystal also gave diffraction spots and some other high-spacing rings. These diffraction spots corresponded to the strongest spots from

a yellow, poorly-shaped octahedron from the Birkbeck College sample and from a white transparent crystal from the University College batch, neither of them diamond. The strong spots from all these single crystals fitted the (0002) line of graphite at 3·4 Å., but the remaining spots were not identified and the agreement may be accidental.

The lines from the black matt octahedral crystal were identical with some found from several, but not all, samples of graphite from the Dayton (Ohio) meteorite, which we had obtained from the Smithsonian Institution, U.S.A. The significance of this agreement between the lines from impurities found among the several batches of G.E. diamonds and from meteoritic material is not known. Nor can we tell whether these extraneous materials were introduced as part of the process of manufacture, or during subsequent crushing and sieving of the synthetic diamonds. It is to be hoped that this information will be released eventually.

Swedish Diamonds.

A small sample of ASEA synthetic diamonds was sent to us through the good offices of Professor G. Hägg of Uppsala and by courtesy of Dr. H. Liander, who had announced their manufacture in an article in the 'ASEA Journal' (1955). The 11-year project had cost about 2 million marks and its success in February 1953 was due to the collaboration of Messrs. Liander, Liljeblad, Lundblad, v.Platen, and Wallin. Temperatures and pressures of up to 3000° C. and 70 000 kg./cm.^{2} were attained. The 12 000-ton press used was made of a new material (unspecified) and was of surprising capacity (500 c.c.). These diamonds are not yet on the market, but further experiments are being made.

The material sent us was not sufficient for a spectrographic analysis, and the individual crystals in our specimen were even smaller and more badly-shaped than those from the G.E. research laboratories. A random sample was formed into a 'powder' specimen and stationary, rotation, and oscillation photographs were taken of it. These showed the presence of a number of very small individual grains of diamond and a few additional powder lines or sections of lines not due to diamond. Separate grains were then set up for X-ray studies.

These separate diamond grains showed, in general, no sharp diffraction spots or lines other than those of diamond itself. It is clear that they do not contain Ni or any other crystallized inclusions. The possibility of impurity atoms within the diamonds is not eliminated by this method,

but if so these are not in the form of even tiny crystals either aligned or not aligned with the diamond. There is no general fluorescent scattering such as would be expected with Cu-$K\alpha$ if iron were present (pl. X, figs. 3–5).

A small portion of the ASEA sample was cleaned with concentrated nitric and sulphuric acids. A subsequent 'powder' photograph still showed some extraneous powder lines which were subsequently identified as due to platinum, present as blackish grains in the specimen. The lines are sharp but fade off in intensity towards the back-reflection region, indicating that the Pt crystallites are large but with a considerable degree of static disorder. It was at first supposed that the Pt (lattice constant 3·92 Å.) might have been introduced as a catalyst in the original matrix and partially recrystallized subsequently. In a communication from Dr. Liander, however, he says: 'This platinum was not intentionally used as a catalyst. In some experiments we have used platinum-canned capsules for the containing of certain reaction mixtures. At the high temperature the platinum has melted and traces of platinum may, therefore, have been attached to these diamonds.'

Laue photographs of G.E. and ASEA diamonds gave no extra spots or streaks typical of 'type I' diamonds (cf. Lonsdale, 1942).

Hannay Diamonds.

It seemed of interest to take a random rotation (Cu-$K\alpha$) photograph of one of the Hannay diamonds (F. A. Bannister and K. Lonsdale, 1943) to see whether any crystalline inclusions were present that would indicate that it must have been made in the laboratory. Pl. X, fig. 6, shows such a photograph. No trace of Ni lines can be seen, although there is one faint powder line present which could not be identified. The X-ray photograph shows that it is a very well-crystallized specimen, in no way distinguishable from a natural diamond. We are inclined to believe that it is a natural and not a synthetic diamond. One point may, however, be made. Hannay asserted that he put some lithium into his mixture. Although no lines due to lithium can be seen, it is a fact that lithium does have a lattice constant (3·509 Å.) close to that of diamond, and in spite of its being a body-centred and not a face-centred cubic structure, it might act as a substrate for the growth of diamond. Hannay, of course, could not possibly have been aware of this similarity of lattice constants, which was not discovered until many years after his death.

Appendix.

Spectrographic analyses have been carried out as follows:

A sample sent to us directly from the G.E. Research Laboratories, Schenectady, N.Y., was divided into three parts. One part was retained for X-ray analysis. Two samples, one larger than the other, were sent for spectrographic analysis to the Research Department of Messrs. Johnson Matthey and Co. Ltd., who supplied the information given in columns 1 and 2 of table III.

Two samples were given to us by Mr. P. Neven, Director of the Impregnated Diamond Products Ltd. One of these had been treated with a mixture of concentrated nitric and hydrochloric acids, followed by molten potassium hydroxide with the addition of potassium nitrate at about 400° C., and finally by a second treatment with aqua regia. As a result of this treatment there was a loss of weight of up to 1·5 %, but with no noticeable change in the dirty greenish-grey colour of the sample. The spectrographic analysis of a random portion of this sample is given in column 3 of table III. The second sample was entirely untreated, and the analysis of a random portion of this is given in column 4.

Table III. Spectrographic analyses of synthetic diamond.

	1. Larger sample from G.E. lab.	2. Smaller sample from G.E. lab.	3. Cleaned G.E. industrial	4. Original G.E. industrial
Nickel	0·2%	0·2%	3(±1)%	1(±0·3)%
Silicon	0·05	0·1	0·005	4(±1)
Aluminium ...	0·07	0·07	—	0·05
Iron	0·002	0·05	0·0002	0·0005
Manganese ...	0·003	0·005	0·002	0·002
Magnesium ...	0·002	0·01	0·005	0·002
Sodium	0·002	0·005	0·003	—
Potassium ...	0·005	0·005	—	—
Titanium ...	0·003	0·005	—	—
Calcium	0·002	0·01	0·005	0·003
Chromium ...	—	0·005	—	—
Copper	0·0002	0·004	0·0001	0·003
Boron	—	0·0005	—	—
Thallium ...	—	—	0·05	—
Silver	—	—	—	0·003

The following elements were tested for (if not mentioned in the table they were not present in detectable amounts): Ag, Al, As, Au, B, Ba, Be, Bi, Ca, Cd, Co, Cr, Cs, Cu, Fe, Ga, Ge, Hg, In, Ir, K, Li, Mg, Mn, Mo, Na, Ni, P, Pb, Pd, Pt, Rb, Rh, Ru, Sb, Si, Sn, Sr, Te, Th, Ti, V, W, Zn, Zr.

The fact that specimens 1 and 2 differ, although taken from the same sample, is a measure of the variability of the material. This is also shown by specimens 3 and 4, since one would expect that 4 should contain all the elements found in 3, as well as those (mainly Si) removed by the treatment, whereas thallium and sodium were found in the cleaned, but not in the uncleaned samples.

Some of the impurities found may have been added accidentally after the initia manufacturing process was completed. Our X-ray diffraction studies, for example, have shown the presence of some grains of copper as foreign bodies.

Acknowledgements. Our thanks are due to all those mentioned in the paper who supplied us with specimens or information; also to Drs. R. Mason and S. Chandrasekhar, who assisted with experimental measurements, and to Dr. H. Morrogh, who has made various nickel carbides for X-ray examination.

References.

Anderson (B. W.), 1958. Gemmologist, vol. 27, p. 59.
Bacon (G. E.), 1950. Acta Cryst., vol. 3, p. 320.
Bannister (F. A.) and Lonsdale (K.), 1943. Min. Mag., vol. 26, p. 315.
Bernier (R.), 1951. Ann. Chim., vol. 6, p. 104.
Bundy (F. P.), Hall (H. T.), Marshall (A. L.), Nerad (H. J.), and Strong (H. M.), 1955. Man-Made Diamonds. G.E. Research Information Services.
—— —— Strong (H. M.), and Wentorf (R. H., *Jnr.*), 1955. Nature, vol. 186, p. 51.
Grenville-Wells (H. J.), 1952. Min. Mag., vol. 29, p. 803.
—— and Lonsdale (K.), 1958*a*. Nature, vol. 181, p. 758.
—— —— 1958*b*. Symposium on Crystal Physics. Nat. Inst. Sci. India (in the press).
Hall (H. T.), 1956. Article on pp. 161–166 of the Symposium: High Temperature—A Tool for the Future. Stanford Research Institute, California, U.S.A.
—— 1958. Science, vol. 128, p. 445.
Liander (H.), 1955. ASEA Journal, p. 97.
Lipson (H.) and Stokes (A. R.), 1942. Nature, vol. 149, p. 328.
Lonsdale (K.), 1942. Proc. Roy. Soc., ser. A, vol. 179, p. 315.
Nath (N.), 1935. Proc. Indian Acad. Sci., vol. 2, p. 143.
Slawson (C. B.), 1957. Amer. Min., vol. 42, p. 299.

Explanation of Plates IX and X.

Plate IX. Fig. 1. (*Top, left.*) Natural diamond twins rotating about $[01\bar{1}]$ and $[4\bar{1}1]$ (slightly mis-set). Cu-K filtered radiation. Compare table II.

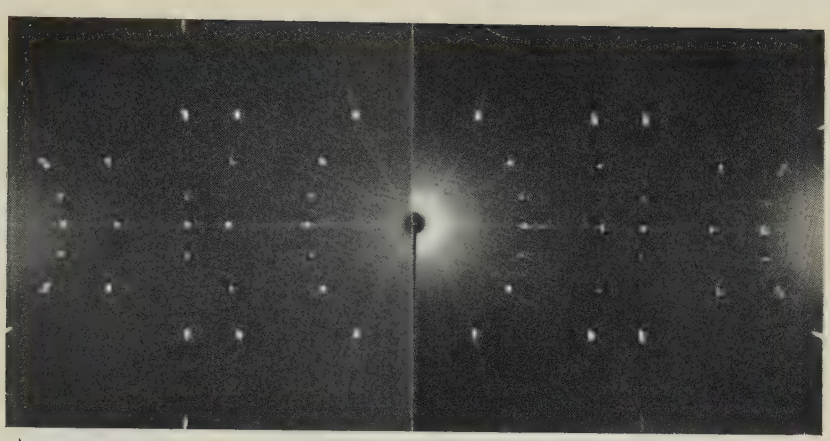
Fig. 2. (*Top, right.*) G.E. green diamond twins rotating about $[01\bar{1}]$ and $[4\bar{1}1]$ (slightly mis-set). Cu-K filtered radiation. Note satellite spots, especially those corresponding to 200, 002, and 020 on zero, first, and third layer lines, and their complete parallelism with the diamond matrix structure.

Fig. 3. (*Middle, left.*) Another G.E. diamond with weak twin spots. Strong Ni satellite spots at 200 and 020. The 002 twin Ni satellite spots are visible on the film but too weak to reproduce. Note the 222 doublet due to diamond and Ni (the latter being the stronger), and the 133 satellite.

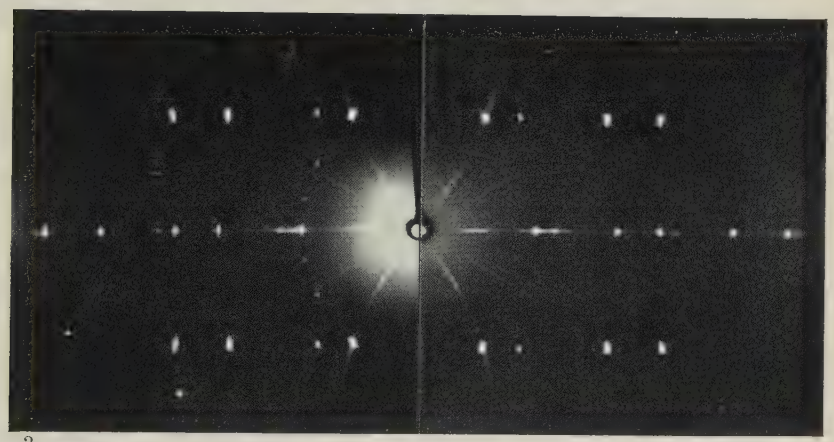
Fig. 4. (*Middle, right.*) A single G.E. diamond with a strong single Ni satellite. Note the 222 doublet, and 133 satellite.

Fig. 5. (*Bottom.*) Upper half, left: Diamond powder + 0·3 % Ni powder. Upper half, right: Diamond powder. Lower half: Ni powder. All taken with Cu-K filtered radiation: showing how Ni would give clear 200 and 420 reflections and satellites on the high angle side of all diamond reflections.

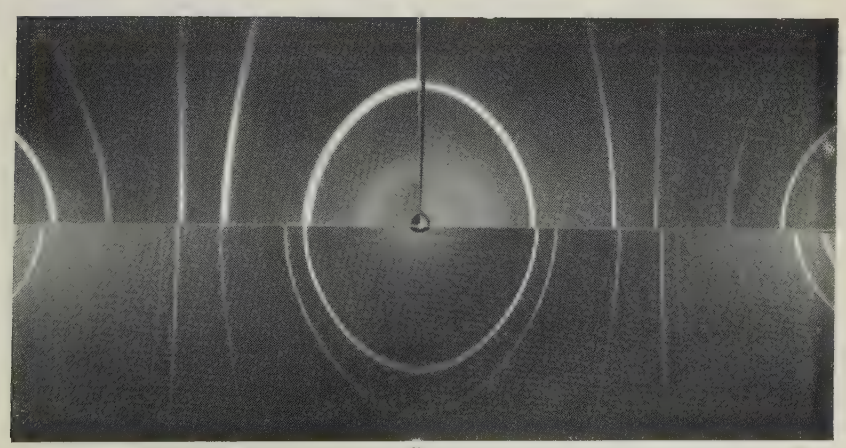
Plate X. Fig. 1. (*Top, left.*) Random rotation, with unfiltered Cu-K radiation, of grey-black granular G.E. diamond (part of specimen returned after spectrographic analysis). The Ni satellite spots are fairly strong and show some spreading along arcs. The 200 satellite spots show no $\Delta\theta$ broadening but the 420 satellite spots are broadened, giving evidence of strain. A powder inclusion gives a strong 3·08 Å. line.



1 2

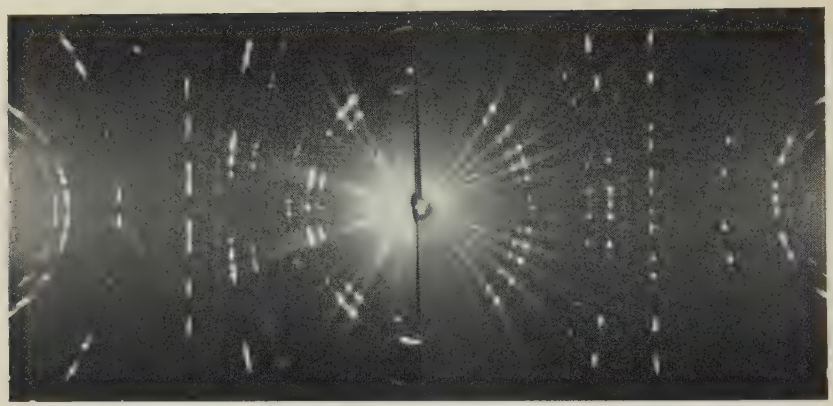


3 4

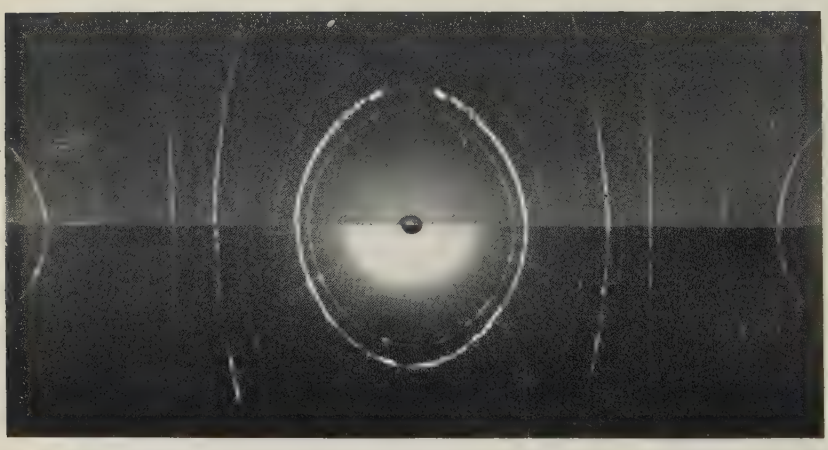


5

K. Lonsdale, H. J. Milledge, and E. Nave: Synthetic Diamond

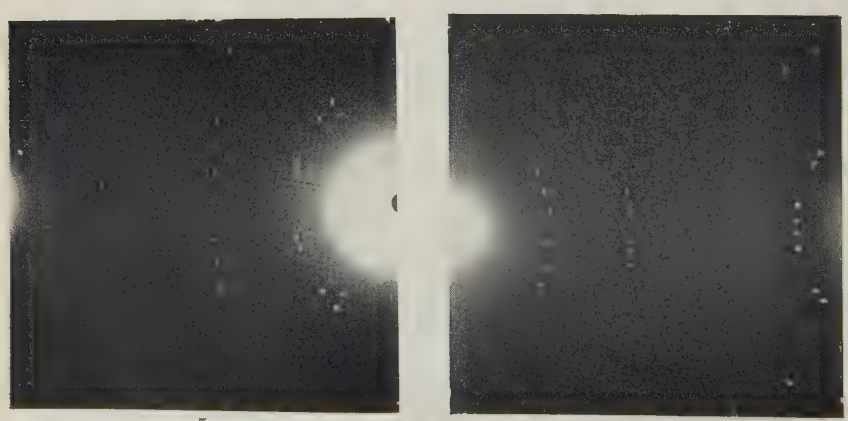


1 2



3

4



5 6

K. LONSDALE, H. J. MILLEDGE, AND E. NAVE: SYNTHETIC DIAMOND

FIG. 2. (*Top, right.*) The same powder inclusion line is found from another G.E. diamond, which consists of black parallel cubes. The Ni satellite spots do not spread along arcs.

FIG. 3. (*Middle, upper half.*) G.E. diamond, showing 200 Ni satellite lines.

FIG. 4. (*Middle, lower half.*) ASEA diamond; no satellite. Both specimens consist of randomly-orientated crystallites.

FIG. 5. (*Bottom, left.*) Random rotation of ASEA single diamond, Cu-*K* radiation.

FIG. 6. (*Bottom, right.*) Hannay diamond. Single crystal with no satellite, though the film shows a few weak powder lines which do not reproduce.

The amygdale minerals in the Tertiary lavas of Ireland. II. The distribution of gmelinite.

By George P. L. Walker, M.Sc., Ph.D., F.G.S.

Department of Geology, Imperial College, London.

[Read 4 June 1959.]

Summary. Mapping of the distribution of zeolites in the Antrim basalts has revealed that gmelinite is confined to a narrow zone along the eastern seaboard of Antrim. This zone comprises less than 1% of the area of the basalts, but in it gmelinite is extremely abundant. The gmelinite zone is superimposed upon the lava pile, and the minerals in it are clearly later than the lavas; it is in the form of a dome, elongated north-west and traceable for 18 or 24 miles in this direction, and is found in the basal parts of the lava pile, locally reaching a height of some 500 feet in the pile. The zone may be related in its position and origin to the Islandmagee fault zone. Gmelinite frequently forms a parallel overgrowth on chabazite. Superimposed upon the southern part of the gmelinite zone is a zone of late chabazite and heulandite containing, *inter alia*, parallel overgrowths of late chabazite on simple or twinned gmelinite.

A STUDY of the amygdale minerals in the Tertiary basalts of the Garron Plateau area (Walker, 1951) showed that the zeolites and the crystal habits of chabazite have a regular vertical distribution in the lavas. The present paper concerns the distribution of the zeolite gmelinite. This mineral is confined to a narrow belt of lavas along the eastern seaboard of Antrim, referred to in this paper as the gmelinite zone, but within this restricted zone it is extremely abundant. Superimposed upon the gmelinite zone in its southern part, and encroaching also upon the other adjacent zeolite zones, is a zone of late chabazite and heulandite, which is also described below.

In what follows, after the minerals have been described, the shape and relations of the gmelinite zone are considered, and the conclusons to be drawn from this study reviewed.

Previous work.

Gmelinite has long been known to occur in the east of Antrim, and most mineral collections contain specimens. Bryce referred to gmelinite as occurring at Glenarm and in Islandmagee in 1833, and in 1837

Brooke described crystals of chabazite from Glenarm with a parallel overgrowth of gmelinite. Portlock, in his excellent summary of the amygdale minerals of the Antrim Basalts, published in his memoir of 1843, gave as localities Islandmagee, Black Head, Glenarm, Portrush, and several places in County Londonderry.[1] This list was repeated by Greg and Lettsom (1858) and Hintze (1897).

The only work published since then is the paper by Smith and Ashcroft (1916) dealing, among other topics, with chabazite and gmelinite from Whitehead, Islandmagee. Apart from this paper, very little has been added to the knowledge of the minerals of the gmelinite zone since the time of Bryce and Brooke over a century ago.

The only other British record of gmelinite is that by Heddle (1901) from Talisker, Skye, and a specimen from this locality in the Royal Scottish Museum, Edinburgh, is unquestionably gmelinite. Farther afield, gmelinite is much more restricted in occurrence than many other zeolites. Hintze (1897) listed as localities Skye, Vicenza (Italy), Cyprus, Andreasberg (Harz), Austria, Nova Scotia, New Jersey, Mexico, and Victoria (Australia). Since then gmelinite has been recorded from a few other localities (Sicily, Dana 6th edn., app. ii, p. 46; S. Rhodesia, M.A. **6**–159; Russia, M.A. **2**–299, **8**–298).

Geological environment.

The best exposures of gmelinite-bearing basalts in Antrim are in the outliers on the peninsula of Islandmagee, where there are excellent cliff-sections around the north and east coasts. Unfortunately there are very few exposures inland. Other exposures of gmelinite-bearing basalts are found at Whitehead; Black Cave, north of Larne; and along the basalt escarpment, often precipitous, east and south-east of Glenarm, where good specimens may be collected from screes and slipped debris.

In these exposures a thickness of up to about 600 feet of basalt lavas is exposed, embracing in north Islandmagee and near Glenarm the whole of the Lower Basalts (Cole, 1912) and the lower part of the Upper Basalts. Both series of lavas are composed almost exclusively of olivine-basalts (Tomkeieff, 1934), with some picrite-basalts. In Islandmagee they include the best exposures in Antrim of thin pahoehoe flow-units, with pipe-amygdales at the base and ropy tops. Every flow contains

[1] Portlock's identification of gmelinite at Portrush and localities in Co. Londonderry is doubtful; the present writer has not seen gmelinite in the field there or in mineral collections. Phacolite, which is sometimes pink and closely resembles gmelinite, was probably misidentified as gmelinite.

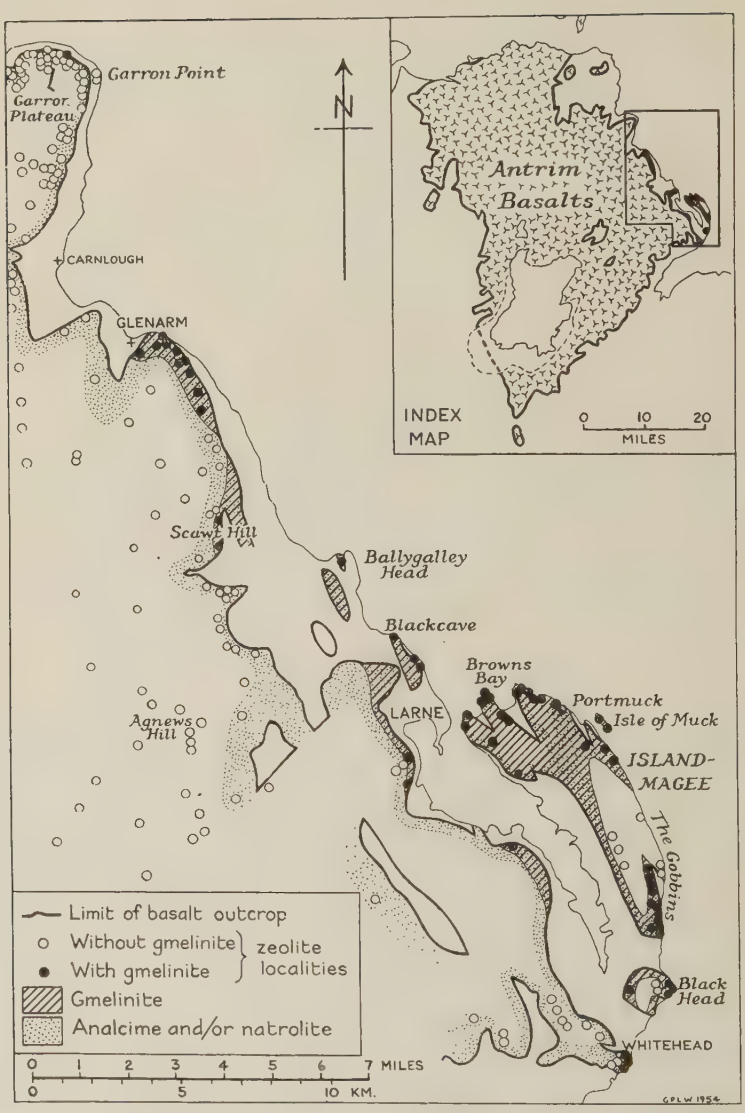


FIG. 1. Map showing the distribution of gmelinite in the Antrim Basalts.

zeolites and other secondary minerals such as calcite in amygdales and also in the body of the rock, and these minerals form probably around 10 % of the basalt pile.

The minerals.

Gmelinite is one of a rich suite of secondary minerals, many of which accompany it in the same cavity, and the following list includes most of the minerals that are found:

Gmelinite. The index mineral of the gmelinite zone, it is perhaps the most abundant zeolite in it. It occurs as well-formed crystals up to about 1 cm. but normally several mm. in diameter. Only rarely does the mineral form radiating aggregates, unlike many of the other zeolites. It is usually pink in colour, and is optically uniaxial negative, with mean refractive index ranging from 1·470 to 1·485 and birefringence from about 0·002 to 0·005.

The crystals are combinations of the forms $r\{10\bar{1}1\}$, $\rho\{01\bar{1}1\}$, $m\{10\bar{1}0\}$, and $c\{0001\}$. The following five main habits may be distinguished (fig. 2):

Rhombohedral habit: forms m, r, and ρ, with r and ρ unequally developed. There is usually a curved concave vicinal strip in the zone $r\rho$ along the terminal edges. Also commonly present are vicinal faces in the zones rm, ρm, and mm.

Pyramidal habit: forms m, r, and ρ, with r and ρ equally developed. Vicinal faces in the zone $r\rho$ are usually absent or inconspicuous. This is the most common habit.

Tabular habit: as pyramidal habit, but with c present. As c increases in relative size, the crystals become thin and platy. The faces of c are rarely bright and plane. The crystals usually have a basal parting.

Prismatic habit: as pyramidal habit, but elongated along the c-axis with prominent m faces which are usually strongly striated horizontally.

Twinned habit: crystals, generally of tabular habit, but sometimes pyramidal, showing twinning on r in which several individuals are united to form a twin of unusual complexity (Smith and Ashcroft, 1916, figs. 9, 10).

Chabazite occurs as well-formed crystals usually up to 5 mm. in diameter and often pink in colour, like the gmelinite. The habit is 2 or 3, following the nomenclature used by Walker (1951), and only rarely are forms other than $\{10\bar{1}1\}$ prominent, except in the late chabazite zone.

Optically the chabazite is negative, and either uniaxial or biaxial with small optic axial angle. Refractive indices measured range from 1·470, rather lower than that of chabazite found elsewhere in the Antrim Basalts, to about 1·494, with a birefringence of the order of 0·003.

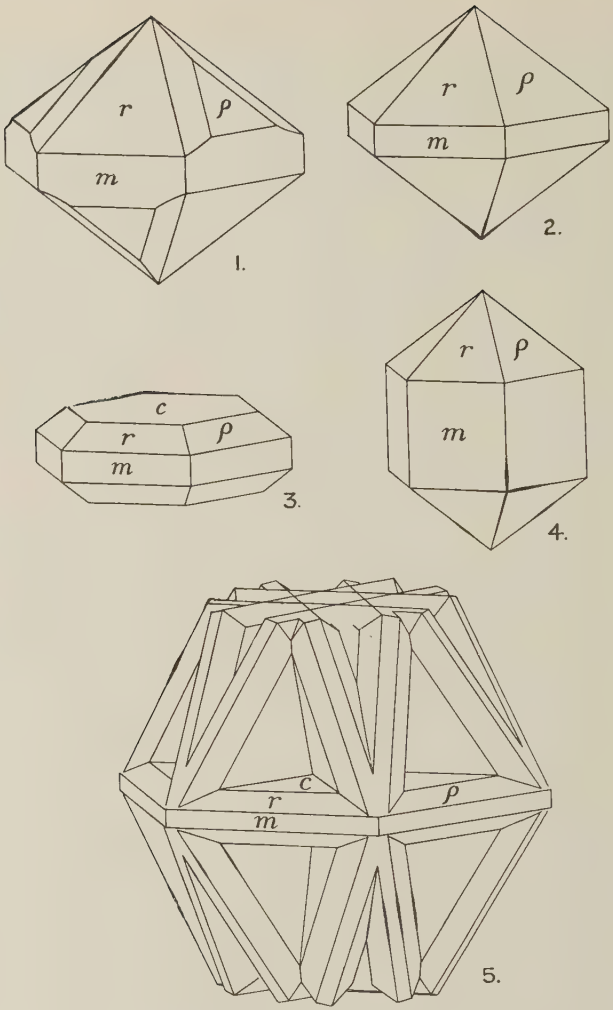


FIG. 2. The five crystal habits of gmelinite in Antrim. Forms present: $r\{10\bar{1}1\}$, $\rho\{01\bar{1}1\}$, $m\{10\bar{1}0\}$, and c $\{0001\}$. The drawing of the twin-crystal is idealized, and for clarity some edges are omitted.

Levyne is quite common and widespread, as small platy crystals which tend to occur alone in a cavity, although they may at times be associated with other zeolites. It is often pink in colour. Levyne from the gmelinite zone is uniaxial negative, with mean refractive index 1·489 to 1·495, and birefringence about 0·002–0·006. The levyne commonly shows the

oriented overgrowth of a fibrous mineral, probably an alteration product, as is common elsewhere in Antrim (Walker, 1951).

Analcime is abundant in much of the gmelinite zone, and differs in three ways from that found elsewhere in the Antrim Basalts. It forms small, transparent and glassy crystals rather than the large, dull white crystals which are usual elsewhere. Although the crystals are usually of simple habit, with {211} alone, other forms are sometimes developed; these include {100}, {332}, and rarely {111}, forms which have not been observed elsewhere in the British Tertiary (Heddle, 1899). Finally, at a number of localities radiating aggregates of glassy analcime are found, and again these have not been observed elsewhere in Antrim.

Natrolite is fairly common as stout, prismatic crystals terminated by {111}, much stouter than are usual elsewhere in Antrim; the crystals are either in loose aggregates or closely spaced in compact masses with radiating structure. Perhaps the best natrolite is seen in the quarries at Whitehead, now unfortunately disused, and at Glenarm. Optically it is biaxial positive, with refractive indices varying little from γ 1·488, β 1·478, and α 1·476.

Thomsonite is represented by the variety faröelite, as more or less hemispherical radiating aggregates of closely-packed acicular crystals elongated parallel to *c*. It is either white or pink. Although quite common, thomsonite is much less abundant in the gmelinite zone than in most of the remainder of the Antrim Basalts, and much of it may have been converted to other zeolites. Optically it is biaxial positive, with β ranging from 1·515 to 1·526, and γ 1·523 to 1·535.

Phillipsite, although fairly widespread, is seldom abundant. Perhaps, the best material may be collected from the coast south-east of Portmuck, Islandmagee. The crystals are normally pink, deeper in colour than gmelinite, and are nearly always in radiating aggregates; when in single crystals the habit sometimes closely resembles a rhombic dodecahedron.

Calcite is extremely abundant throughout the gmelinite zone, in cavities and in veins probably related to the faulting. *Aragonite* is present in much smaller amount, in radiating aggregates. *Quartz* and *chalcedony* have not been observed, and neither have *apophyllite* and *gyrolite*, although Portlock (1843) recorded apophyllite from Islandmagee. Bryce (1833) recorded *gismondine* from the Gobbins, Islandmagee, but the mineral referred to is almost certainly the mixture of chabazite and heulandite found in the late chabazite–heulandite zone. Specimens of this aggregate, labelled gismondine, are often found in the

older mineral collections. Of the rarer minerals, *scolecite* and *mesolite* are sometimes encountered, usually as chalk-like matted aggregates of minute fibres, and *native copper* and *malachite* have been found by the writer in minute amounts at several places. Exceptional are the amygdale minerals near Portmuck, Islandmagee, developed near intrusive dolerite dykes (Walker, 1948), which include *andradite*, *apatite*, *magnetite*, *diopside*, *ilmenite*, and *sphene*.

The order of crystallization.

In the gmelinite zone most of the amygdale minerals listed above may coexist at the same locality, and many of them in the same cavity, although there is a tendency for levyne and to a lesser extent analcime and phillipsite to occur alone in a cavity. The limited available evidence points to levyne and phillipsite as the earliest zeolites to crystallize, levyne preceding phillipsite; they were probably followed closely by chabazite and thomsonite.

The more sodic zeolites, gmelinite, analcime, and natrolite, came later. Gmelinite unquestionably follows chabazite, for at every locality where the two are associated –and they are associated at most localities —some of the gmelinite forms minute crystals in a parallel overgrowth on chabazite, as figured by Brooke (1837). Analcime and natrolite are later than gmelinite.

Calcite is very abundant throughout the zone, especially so near faults, and it may be either earlier or later than the zeolites. The earliest calcite is often in small hemispherical radiating aggregates. Aragonite tends to fill the amygdale in which it occurs, but whenever zeolites are seen associated with it they are of later formation. Much of the aragonite has subsequently been replaced by a sugar-grained aggregate of calcite.

Discussion of the late chabazite–heulandite zone is deferred (p. 214), but it may be noted that in this zone the two index minerals are later than all the other amygdale minerals described above.

The colour of the minerals.

A striking feature of the gmelinite zone is the pink colour of many of the minerals. Gmelinite is usually pink, and so are the associated chabazite, phillipsite, thomsonite, and levyne, these minerals being almost invariably white in the remainder of the Antrim Basalts outside the gmelinite zone. It would appear that the conditions which were favourable to the formation of gmelinite were also those which promoted

a pink coloration of the minerals. The minerals of the late chabazite–heulandite zone are, however, white.

It is interesting to note that, at Garron Point, Glenarm, and a few places farther south the extreme edge of the gmelinite zone is not quite coincident with the edge of the zone of coloration, and a little colourless gmelinite appears. The gmelinite collected by Heddle at Talisker, Skye, is similarly colourless.

The shape of the gmelinite zone.

Gmelinite has been found by the writer at some sixty localities in Antrim. Its western limit is marked by a line trending north-west that can be traced for 18 miles from Whitehead past Larne and Scawt Hill to near Glenarm. This line is extended to 24 miles if account is taken of the occurrence of gmelinite at a single locality near Garron Point. The eastern limit of gmelinite in the lavas is nowhere seen, lying somewhere off the coast, and the greatest known width of the gmelinite zone is $3\frac{1}{2}$ miles in north Islandmagee.

In the lower 50 to 100 feet of the lava pile within the limits of the gmelinite zone there are no localities at which gmelinite is not present. The distribution at higher levels in the lavas is much more restricted. On the escarpment south of Larne gmelinite is found in quarries at the base of the lavas, but is absent in the large quarries on Carnduff Hill 100 to 200 feet higher up. At Whitehead the quarries and coast exposures in the lowermost 100 feet of lavas are rich in gmelinite, but this mineral is not found in road cuttings at the top of the headland some 200 feet above the base of the lavas. A similar relationship is found in the southern half of Islandmagee: the Black Head outlier does not bear gmelinite in the several small openings at the top of the cliffs near the lighthouse, but gmelinite is abundant lower down along the shore, and at the base of the lavas on the west side of the outlier. At the Gobbins, gmelinite is abundant in the lower 100 feet of lavas but is absent from most of the road cuttings 100 feet or so higher up.

In the southern half of Islandmagee, then, the roof of the gmelinite zone is exposed, and the zone has a thickness of the order of 100 or 200 feet. In the north of Islandmagee, on the other hand, the thickness is much greater, gmelinite being distributed throughout the Lower Basalts and in at least the lower 100 feet of the Upper Basalts, near Brown's Bay. That is, the gmelinite zone attains a height of at least 500 feet in the lava succession.

The conclusion to be drawn on the shape of the zone is that it forms

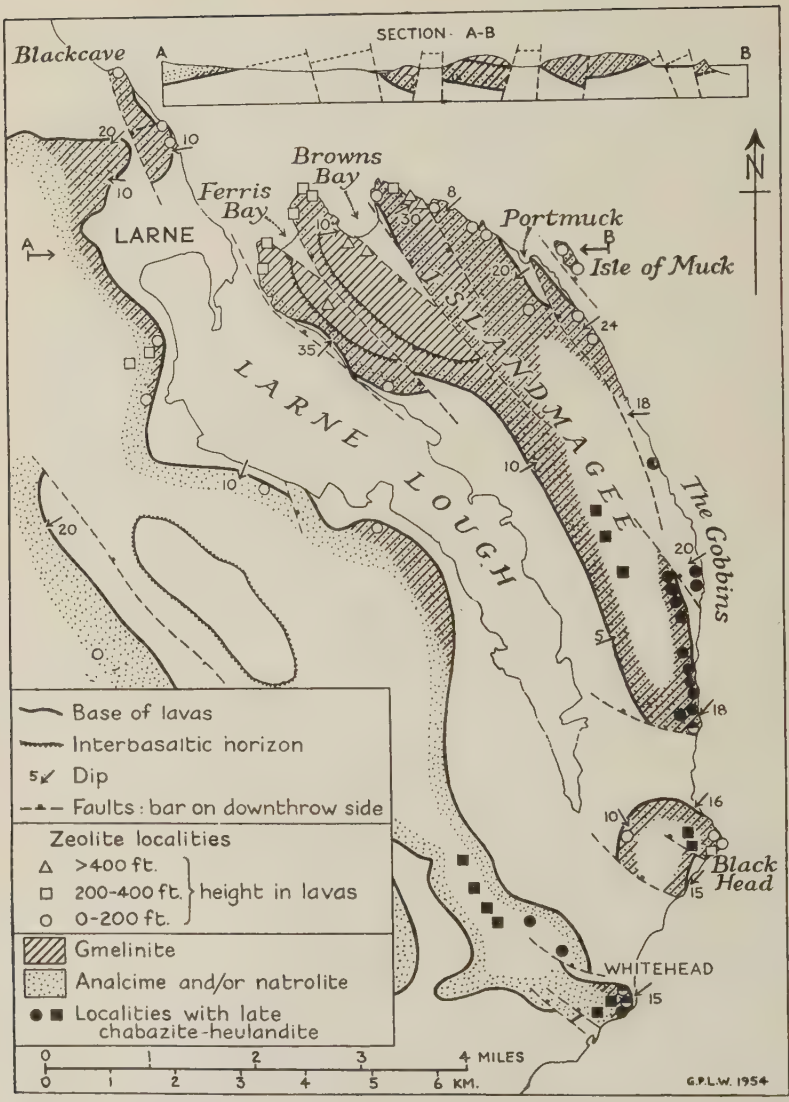


FIG. 3. Geological map of Islandmagee showing the distribution of amygdale minerals. The known faults of the Islandmagee fault zone are indicated on the map and section above.

an elongated dome trending north-west, less than 100 feet thick along the western margin but up to 200 feet thick at Black Head and at least 500 feet in north Islandmagee, where the zone has its greatest known width. The greatest known lateral extent of the zone is at the base of the lava pile.

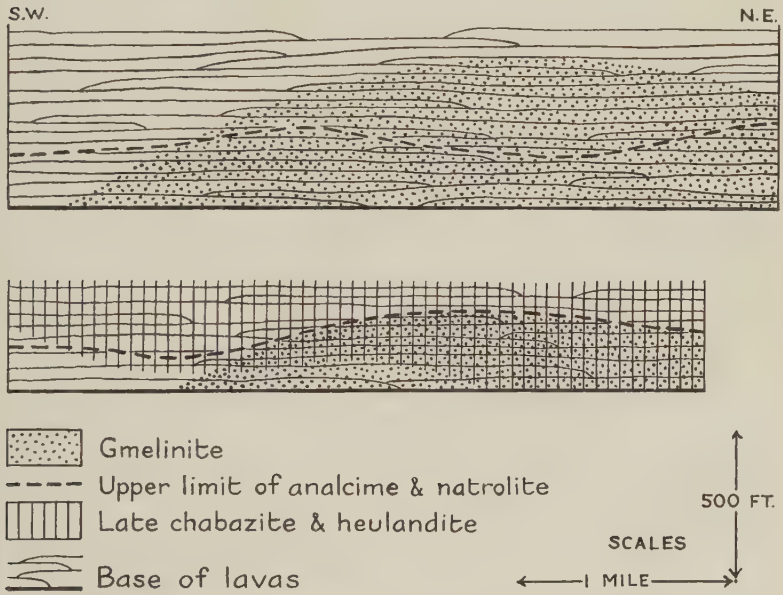


FIG. 4. Diagrammatic sections across the gmelinite zone: above, in north Islandmagee; below, in south Islandmagee. The lavas are indicated schematically.

The distribution of gmelinite habits.

Following the success achieved in mapping the distribution of chabazite habits in Antrim (Walker, 1951), an attempt was made to map the distribution of gmelinite habits. Unfortunately the nature of the exposures makes this very difficult, for nowhere in the gmelinite zone are exposures found that are more than 200 or 300 feet high, and exposures inland from the coastal cliffs are almost non-existent. The extensive faulting adds to the difficulty.

The only relationship that has been established is the marked tendency for crystals of platy, twinned, and prismatic habits to be confined to the upper and lateral marginal parts of the gmelinite zone, the pyramidal and rhombohedral habits being characteristic of the remainder of the

zone. Thus gmelinite of prismatic habit is developed along the edge of the zone at Garron Point, Glenarm, near Scawt Hill, and to a limited extent at Whitehead, and the platy and twinned habits at Whitehead and where the gmelinite zone is thin north of Black Head and at the Gobbins.

The age and origin of the gmelinite zone.

The gmelinite zone is clearly superimposed upon the lava pile, and gmelinite must have been formed at some time after the eruption of the Lower Basalts and at least part of the Upper Basalts.

When one attempts to correlate the gmelinite zone with some geological feature, it is seen to be roughly coincident with the Islandmagee fault zone, a fracture belt without known parallel elsewhere in the Antrim Basalts. In Islandmagee and in adjacent parts of the mainland the basalt lavas are cut by a set of large north-west-trending faults into narrow strips tilted in different directions. The gmelinite zone attains its greatest known thickness where the fracturing is most intense, in the area of the horsts of Brown's Bay and Ferris Bay in the north of Islandmagee.

Most of the faults in the Islandmagee fault zone are not indicated on published geological maps; those known to the writer have been inserted on the accompanying map, fig. 3. Collectively they have a throw of over 3000 feet (not all throw in the same direction) and are probably related to the formation of the North Channel between Ireland and Scotland.

One is tempted to postulate the formation of gmelinite through the agency of hot solutions rising along the faults, but the gmelinite zone appears to have been itself shifted by some of these faults. The Gobbins fault appears to have downfaulted the gmelinite zone to below sea-level on the east side, whereas gmelinite is found in the lower 100 feet of lavas on the west side. Along the Castlerobin fault in north Islandmagee, which has a downthrow of the order of 300 or 400 feet west, gmelinite is of different habit on opposite sides of the fault, and is much less abundant on the downthrow side.

Although sought for, only a single convincing example of a bedded floor to an amygdale was found. In this example the cavity was floored with red zeolitic material and its upper surfaces lined with gmelinite. It was found on the western shore of Brown's Bay in a fault block tilted at 10°, the bedded floor being likewise tilted and showing that tilting postdated the formation of the bedded deposit. It is assumed that tilting and faulting were essentially contemporaneous.

The limited available evidence would appear to indicate that the bulk of the faulting postdated zeolitization, but the areal relationship of the gmelinite zone and the Islandmagee fault zone seems too close to be coincidental. Perhaps the faults were initiated before gmelinite crystallized, although the larger part of the faulting took place later.

Zeolitization by dykes.

In three sea-stacks ½ mile north-west of Portmuck, Islandmagee, a large dyke of olivine-dolerite cuts a succession of thin flows of olivine-basalt and has caused considerable alteration to a distance of some 15 feet from the contact (Walker, 1948). This alteration includes extensive zeolitization of the lavas. Amygdales and veins in the lavas near the dyke contain zeolites accompanied by andradite, apatite, magnetite, soda-diopside, ilmenite, sphene, and calcite. Thomsonite and natrolite are very much more abundant near the dyke than in the lavas farther away, and similar relationships are found with another (probably related) dyke on the Isle of Muck.

These are clear examples of development of zeolites in the hydrothermal aureoles about a dyke, but the effect is very local. On one of the stacks north-west of Portmuck a small dyke cuts the main dyke and is chilled against it, and its amygdales contain gmelinite and a mineral assemblage similar to that in the lavas in the whole district. One is forced to the conclusion that although some zeolites were formed by dykes at Portmuck, the bulk of the zeolitization and the regional formation of gmelinite in the lavas postdated the intrusion of the dykes.

The relative age of faults and dykes in Islandmagee can be inferred from the following evidence: with the exception of the Gobbins fault, the faults of Islandmagee do not carry dykes despite the easy path they would presumably offer to the passage of basaltic magma; and the dykes in Islandmagee invariably cut the lavas approximately normal to the stratification. In the fault blocks the lavas are sometimes tilted quite steeply, but this relationship still holds, and the dykes must have been tilted with the lavas. This evidence indicates that dyke intrusion antedated faulting and tilting of the lavas.

The distribution of analcime and natrolite.

In the country west of the gmelinite zone, the distribution of amygdale minerals follows the same pattern as that worked out (Walker, 1951) for the Garron Plateau area. Two distinct amygdale mineral assemblages may be mapped; the one, characterized by the abundance of analcime

or natrolite or both, together with other zeolites such as chabazite, forms a nearly continuous zone in the lowermost few hundred feet of the lava pile; the other, characterized by the abundance of chabazite and thomsonite to the exclusion of analcime and natrolite, overlies it. Evidence from the Garron Plateau area indicates that chabazite and thomsonite mostly crystallized before the formation of north-west-trending faults, analcime and natrolite after.

The distribution of analcime, natrolite, chabazite, and thomsonite appears to be exactly the same within the gmelinite zone, analcime or natrolite or both being encountered almost everywhere in the lowest few hundred feet of the lavas but not higher up in the lava pile in northern and central Islandmagee. The distribution of analcime and natrolite thus appears to be completely unrelated to that of gmelinite, and both of these minerals are later than gmelinite.

Late chabazite and heulandite.

As noted above, the crystallization of chabazite before gmelinite is well established throughout the gmelinite zone. In the southern half of Islandmagee and at Whitehead, however, there has manifestly been a second period of deposition of chabazite, of different character and appearance to the early chabazite, and closely associated with fine-grained granular heulandite. So distinctive is this second generation that the area of lavas in which it occurs has been mapped and is referred to as the late chabazite–heulandite zone.

The late chabazite–heulandite zone is notable for the remarkable overgrowths formed by the late zeolites. The following effects have been noted:

Fine-grained, granular aggregates of late chabazite and heulandite form a layer up to several mm. thick on the pre-existing minerals; on needles of natrolite, which are nearly always replaced by the late zeolites; on slender scalenohedra of calcite;[1] on hemispherical aggregates of thomsonite, the earlier mineral being often altered to a white chalky substance or replaced by chabazite and heulandite; and on crystals of chabazite, gmelinite, and levyne. The best examples of these overgrowths are found west of the two quarries at Whitehead and in landslips south of the Gobbins.

Late chabazite is commonly observed as a parallel overgrowth on early chabazite. Sometimes the overgrowth is thickest on the lateral edges and corners, as in a small quarry west of Whitehead town. The

[1] A scalenohedron near $\{13.11.\overline{24}.2\}$ is particularly characteristic.

habit of the late chabazite is higher than that of the early (the late has habit 3, the early habit 2, following the notation of Walker, 1951) and as a result little facets of $\{01\bar{1}2\}$ and $\{02\bar{2}1\}$ appear on crystals which originally showed $\{10\bar{1}1\}$ alone.

Late chabazite forms a parallel overgrowth on gmelinite, and usually replaces it as well.

The parallel overgrowths of late chabazite on gmelinite are particularly interesting. When the layer of chabazite is thin or discontinuous, crystals which show faces of both chabazite and gmelinite are the result, and there is little doubt that some crystals described by Smith and Ashcroft (1916) from Whitehead were of this type. Occasionally the overgrowth of late chabazite is clearly visible in hand specimens.

More commonly the late chabazite forms a thick and continuous parallel overgrowth on the gmelinite and largely or completely masks the faces of the earlier mineral, although the general shape of the gmelinite is retained. In nearly all cases the underlying gmelinite has been converted to chabazite.

Typical crystals of this second type from the landslips on the coast south of the Gobbins have the overall shape of tabular gmelinite but with the prism and rhombohedron faces coated with chabazite in parallel orientation, and with $\{0001\}$ coated with granular heulandite. The gmelinite has been converted into chabazite, with loss of its presumed original pink colour, but the basal parting which is so characteristic of gmelinite of tabular habit is preserved as thin air films, imparting a pearly lustre to $\{0001\}$ of the paramorph.

The most remarkable crystals are those in which chabazite forms a parallel overgrowth on gmelinite twins, as found in slips south of the Gobbins and also in road cuttings above the entrance to the Gobbins cliff path. Such crystals must be among the most complex known, for seven or more individuals of tabular gmelinite of the original twin are each coated and replaced by a twin-crystal of chabazite, giving a total of 14 or more chabazite individuals. Each chabazite individual has 18 faces—six each of $\{10\bar{1}1\}$, $\{01\bar{1}2\}$, and $\{02\bar{2}1\}$, neglecting vicinal faces—giving a total of 252 faces. Such a crystal gives reflections from the whole complement of 252 faces, apart from the few missing where the crystal is attached to the rock. In such twins, trios of $\{02\bar{2}1\}$ faces of chabazite, grouped around the main triangular re-entrants of the twin, reflect light almost simultaneously.

It has long been known that the axial ratios of chabazite and gmelinite are closely related, and Smith, from the evidence supplied by crystals at

Whitehead, regarded the two minerals as identical. Pirsson (1891) had concluded that the discrepancy in the relation c (chabazite) = 3/2 c (gmelinite) was too great for identity of gmelinite with chabazite, and more recently Bannister (Hey, 1950) has established that they are separate mineral species from X-ray study. That there is a close relationship between the structure of the two minerals is clear from the frequency of parallel overgrowths and paramorphs.

Tomkeieff (1934) has analysed the chabazite, presumably late chabazite, from the Gobbins, and proved it to be rather lower in silica and higher in alumina and soda than most analysed chabazite.

Shape and relations of the late chabazite–heulandite zone.

The late chabazite–heulandite zone embraces the southern half of Islandmagee and the ground near Whitehead. At the Gobbins it extends to the base of the lavas, but at Black Head and Whitehead the base of the zone is 100 feet or more higher. The zone appears to be unrelated to the gmelinite zone, being superimposed upon it and upon the contiguous zeolite assemblage zones west of Whitehead, and there is no evidence that it has been shifted by any of the faults. It does not seem possible at present to relate it to any geological feature or episode.

Conclusions.

Mapping of the distribution of gmelinite has shown clearly its restriction to a zone of small extent in the east of Antrim, a zone the boundaries of which can be accurately delineated and the shape of which can be deduced to be dome-like. The zone is clearly superimposed on the lavas, which establishes the lateness of zeolitization in the volcanic history of the area. The following general sequence of events has been deduced, although it is clear that there must be considerable overlapping of the events:

Eruption of lavas, approx. 500 feet thick (Lower Basalts) and dykes.
Interval; formation of laterite of interbasaltic horizon.
Eruption of lavas (Upper Basalts) and dykes.
Levyne and phillipsite formed, followed by chabazite and thomsonite.
Initiation of faulting in Islandmagee fault zone (?).
Gmelinite formed.
Main period of faulting in Islandmagee fault zone, and tilting of fault blocks.
Analcime and natrolite formed.

Late chabazite and heulandite formed.

Erosion of the area to the present land surface.

It is believed that development of gmelinite in eastern Antrim was related to the fault zone there and perhaps due to heated waters rising along the fractures. When one considers the restricted occurrence of gmelinite, however, it seems that the situation may not have been quite as simple as this, and some unknown factor may have played a part in the development of the mineral.

In Antrim the writer looked for gmelinite at some 700 localities scattered over the 1500 square miles of the Antrim Basalts, but succeeded in finding it at only some 60 localities along the eastern seaboard, covering about 12 square miles of the basalt plateau. Farther afield, there are large areas of basalts containing zeolites but not bearing gmelinite. In Scotland, for example, the apparent absence of gmelinite in the Carboniferous basalts of the Midland Valley and the Tertiary lavas of Mull, and its extreme rarity in Skye are features of which account must be taken in considering its genesis. Gmelinite does not appear to have been recorded at all from the immense piles of zeolite-bearing Tertiary basalts in Iceland, the Faroes, and Greenland, or from many other extensive spreads of basalt lavas.

Clearly gmelinite requires very unusual conditions for its formation in rocks, conditions that in Antrim prevailed only in the extreme east but there succeeded in producing an abundance of the mineral.

References.

Brooke (H. J.), 1837. Phil. Mag., ser. 3, vol. 10, p. 278.

Bryce (J.), 1833. Ibid., vol. 3, p. 86.

Cole (G. A. J.), 1912. The interbasaltic rocks of north-east Ireland. Mem. Geol. Surv. Ireland.

Greg (R. P.) and Lettsom (W. G.), 1858. Manual of the mineralogy of Great Britain and Ireland, pp. 172–176. London.

Heddle (M. F.), 1899. Trans. Edin. Geol. Soc., vol. 7, p. 241.

—— 1901. The mineralogy of Scotland, vol. 2, p. 95. Edinburgh.

Hey (M. H.), 1950. Chemical index of minerals, entry 16.10.26, p. 154. London (British Museum (Natural History)).

Hintze (C.), 1897. Hand. Min., vol. 2, pp. 1787–1788.

Pirsson (L. V.), 1891. Amer. Journ. Sci., ser. 3, vol. 42, p. 59.

Portlock (J. E.), 1843. Report on the geology of the County of Londonderry, p. 221. Geol. Surv., Dublin.

Smith (G. F. H.), Ashcroft (F. N.), and Prior (G. T.), 1916. Min. Mag., vol. 17, pp. 274–304.

Tomkeieff (S. I.), 1934. Geol. Mag., vol. 71, p. 501.

Walker (G. P. L.), 1948. Irish Naturalists' Journ., vol. 9, p. 165.

—— 1951. Min. Mag., vol. 29, p. 773.

Stevensite from the Whin Sill in the region of the North Tyne.

By B. A. O. Randall, B.Sc., Ph.D.

Department of Geology, King's College, Newcastle-upon-Tyne, University of Durham.

[Read 29 January 1959.]

Summary. A stevensite occurring as an alteration product of pectolite in amygdale-like structures within the Whin Sill is described. Data given for the stevensite include optical properties, stain reactions, chemical analysis, X-ray powder data, and D.T.A. curves. The close similarity between many of the properties of sepiolite and stevensite is indicated and D.T.A. is shown to be the most satisfactory method of distinguishing between the two minerals.

STEVENSITE, which occurs as an alteration product of pectolite, was found during an investigation of the Whin Sill in the neighbourhood of Gunnerton, Northumberland. This is the first record of stevensite associated with the Whin Sill.

Stevensite and pectolite are found in large amygdale-like bodies in the upper portion of the sill, but not in the chilled facies near the contact. These bodies resemble pectolite inclusions reported by Tomkeieff (1929) at Caw Burn near Haltwhistle.

At the base of the amygdale-like structure is a layer of rock of coarser grain than the normal sill, which when slightly altered takes on a reddish hue (fig. 1). This 'pegmatitic' rock, which is rich in sulphides and contains over 3 % of datolite by volume (first record of this mineral from the Whin Sill), differs slightly in composition from the normal dolerite.

Pectolite or stevensite forms the main part of the amygdale and invariably has a flattish base but a more undulating upper surface. The minerals show radiating structures, and the centres of growth lie at various points on the periphery. Calcite and datolite are usually present at the upper surface of the pectolite but occasionally are found at the other margins. The top of the amygdale is delineated by a thin layer of chlorite.

Associated with these amygdales are layers of dolerite pegmatite and a second type of amygdale usually only partially filled with smoky quartz and calcite. This second type of amygdale also has a 'pegmatitic'

base, but this base differs in composition from that found below the pectolite structures. Details of these pegmatites will be given in a later paper.

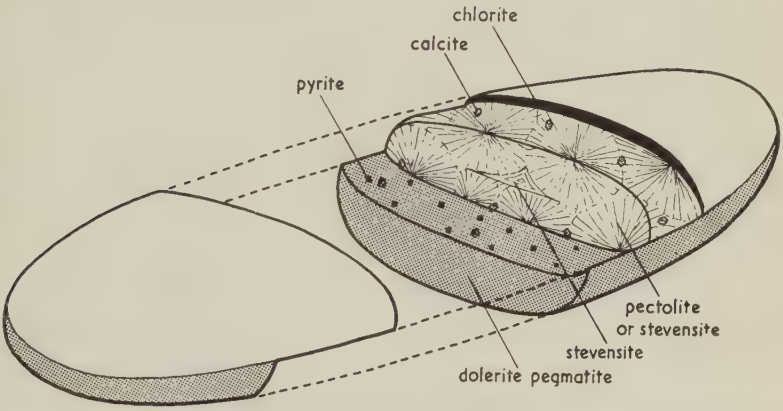


FIG. 1. 'Pull-apart' diagram of 'amygdale-like' body from the Whin Sill, Barrasford quarry, Northumberland.

Identification of the North Tyne mineral as stevensite was hampered by the relatively scarce data available about this mineral, and its close similarity to sepiolite in optical properties, chemical composition, and X-ray data.

The stevensite occurs in two forms: type I is fibrous, and is found as a complete replacement of pectolite, retaining the radiating structure of the original mineral; type II is an amorphous variety, found in small lacunae within the amygdales, encrusting pectolite.

Type I has an earthy lustre and variable colour. The most usual colour is cream, but greenish, brownish, and black varieties have been seen. Colour variations are common even within one radiating orb. Specimens exposed to a dry atmosphere frequently show darkening, become more fragile, and eventually crumble. Type II is usually a bluish-white colour, and possesses a dull lustre.

Optical properties. The two varieties of stevensite have different optical properties. Type I, of fibrous nature, is birefringent, shows straight extinction and positive elongation. The refractive indices of this type fall into two groups, one with $\gamma' = 1{\cdot}543$, $\alpha' = 1{\cdot}537$, $(\gamma' - \alpha') = 0{\cdot}009$ and the other with $\gamma' = 1{\cdot}519$, $\alpha = 1{\cdot}507$, $(\gamma' - \alpha') = 0{\cdot}012$.

The type II stevensite is almost isotropic with $n = 1{\cdot}506$. The two varieties of stevensite from the North Tyne are thus easily distinguished

by use of optical methods, yet they provide identical X-ray powder diffraction patterns.

Stevensite has both birefringent and isotropic forms and in this respect resembles sepiolite. Para-sepiolite is birefringent ($(\gamma-\alpha) = 0{\cdot}008$—$0{\cdot}020$) and has quite variable refractive indices, while β-sepiolite is isotropic ($n = 1{\cdot}517\pm$).

The variation in optical properties of the fibrous forms of both these minerals is probably related to variation in water content.

The *chemical analysis* shown in table I, col. 1, is of fibrous type I stevensite, and was made by Dr. R. A. Chalmers using a modified form of rapid analysis.

Table I. Chemical analyses of stevensite and sepiolite.

	1.	2.	3.
SiO_2	54·5	57·30	54·83
TiO_2	0·02	n.d.	n.d.
Al_2O_3	<0·1	none	0·28
Fe_2O_3	0·34	0·32	0·45
FeO	0·45	none	n.d.
CaO	0·6	0·97	0·55
MgO	26·8	27·47	24·51
MnO	0·41	0·21	n.d.
Na_2O	0·04	0·03	0·35
K_2O	0·09	0·03	0·03
P_2O_5	0·01	n.d.	n.d.
H_2O^+	9·9	7·17	10·74
H_2O^-	6·6	6·69	8·18
	99·9	100·19	99·92

1. Stevensite Type I, North Tyne. Anal. R. A. Chalmers.
2. Stevensite, Springfield, New Jersey. G. T. Faust and K. J. Murata (K. J. Murata anal.), 1954.
3. Sepiolite, Yavapai County, Arizona. A. J. Kauffman, 1943.

The CaO and CaO+MgO were determined by micro methods and the end-point determined spectrophotometrically. Total iron was also done on a micro scale, solution B (Shapiro and Brannock, 1956) being passed through a silver reductor before titration with ceric sulphate.

Table I illustrates the close chemical relationship between stevensite and sepiolite. Since stevensite belongs to the montmorillonite group (Faust and Murata, 1954), the following formula was calculated according to the method outlined by Ross and Hendricks (1945):

$$[Fe^{\cdot\cdot\cdot}_{0\cdot017}\,Fe^{\cdot\cdot}_{0\cdot026}\,Mg_{2\cdot896}\,Mn_{0\cdot026}][Al_{0\cdot075}\,Si_{3\cdot925}]O_{10}[OH]_2$$
$$\downarrow$$
$$R^{\cdot\cdot}_{0\cdot273}$$

Staining tests were carried out on the North Tyne material using methods described by Caillière (1936), Faust (1940), and Faust and Murata (1953). The results of these tests are compared in table II with results published for various clay minerals. When stained with malachite green after acid treatment, the North Tyne reaction was typical of neither stevensite nor montmorillonite (Faust and Murata, 1953), but resembled that of Hector clay.

TABLE II. Stain reactions for stevensite and other clay minerals. Colour in reflected light in parentheses where it differs from that in transmitted light.

	Stevensite N. Tyne, Type I.	Stevensite*	Hector clay†	Montmorillonite‡	Sepiolite‖
Malachite green in nitrobenzene:					
Natural mineral	light green	emerald green	emerald green	—	—
Acid treated	dark emerald green	pale yellow	deep emerald green	yellow–yellow red	—
Alkali treated	dark emerald green	—	deep emerald green	—	—
Methylene blue in nitrobenzene:					
Natural mineral	deep blue green		greenish blue		
Acid treated	deep blue green		greenish blue		
Alkali treated	deep blue green		greenish blue		
Aqueous solutions:					
Malachite green	emerald green (turquoise)				brun violace
Methylene blue	turquoise (bright blue)				bleu violace
Eosin	rose pink (rose madder)				rose saumon
Fluorescein	yellow brown (gamboge)				jaune d'or
Benzidine	Hooker's green	light green powder blue later		purple blue	

*Faust and Murata, 1953.
†Faust, 1940.
‡Mielenz, King, and Schieltz, 1950.
‖Maillière, 1936.

X-ray powder photographs of the North Tyne material were obtained using copper radiation and a 9-cm. diameter camera, which, unfortunately, could not record *d* spacings greater than 10 Å. The *d* spacings recorded are listed in table III, together with data for stevensite and sepiolite. Both types of North Tyne stevensite are shown in this table, and it is interesting to note that there are only negligible differences in the two records. Whilst it is to be remembered that many minerals rich in magnesia have numerous *d* spacings in common, the patterns of sepiolite and stevensite are quite similar. The strong line at 9·78 Å. in the North Tyne material is interesting, because this line is not developed in normal stevensite or sepiolite, although similar lines are reported in both these minerals after heat treatment (Migeon, 1936; Brindley, 1955). The North Tyne material produces lines characteristic

TABLE III. X-ray powder diffraction data for the North Tyne stevensite (1, 2) compared with normal (5) and heated (6) stevensite and with sepiolite (4). Also for a mineral described as sepiolite (3) that appears to be really a stevensite.

1.		2.		3.		4.		5.		6.	
d.	*I.*	*d.*	*I.*	*d.*	*I.*	*d.*	*I.*	*d.*	*I.*	*d.*	*I.*
								24			
						12·15	10	11·2–12·4	10		
9·78	st	9·78	st	10	100	9·8	st*			9·6	m–st
		7·02	vw			7·6	3				
		5·01	w	5	10	5·05	2	4·89–4·96	2	4·8	w
4·52	st	4·53	st	4·52	90	4·60	10	4·54	10	4·5	w
4·29	vw										
3·89	vw					3·82	5				
						3·60	1				
						3·40	2				
3·22	m–st	3·20	st b	3·20	70	3·22	5	3·0–3·5	5	3·18	m–st
3·04	m										
2·87	w										
2·67	w										
2·60	st	2·59	st			2·61	8	2·618	9		
				2·54	90						
2·49	st	2·49	st								
						2·45	7				
2·28	w–m	2·27	w	2·27	20	2·28	6	2·279	2		
2·24	w										
2·09	w–m	2·09	w			2·09	5				
		1·97	vw			1·97	1				
1·91	w–m	1·90	vw	1·92	10					1·92	w b
1·87	w–m					1·88	2				
1·81	vw										
1·72	m	1·72	m	1·72	30	1·71	2	1·725	4		
		1·70	w								
1·61	vw					1·60	4				
						1·56	4				
1·52	vst	1·52	vst	1·52	90	1·52	2	1·520	9		
1·43	vw										
1·37	vw					1·38	2				
						1·36	2				
1·31	st b	1·31	st	1·31	50	1·31	3	1·314	7		
1·26	w	1·26	w–m	1·26	10			1·265	2		
1·24	w									1·22	w b
1·18	vw										
1·14	vw										
1·04	w	1·04	vw					1·047	1		
0·992	m b	0·991	m b	0·993	30			0·994	3		
0·879	m–w	0·879	mb	0·879	30			0·879	3		

1. N. Tyne, Type I, fibrous stevensite.
2. N. Tyne, Type II, stevensite.
3. Stevensite ('sepiolite'). Copt Hill, A.S.T.M. index card no. 2–0048.
4. Sepiolite. Longchambon, 1937.
5. Stevensite. Brindley, 1955.
6. Stevensite heated to 400° C. Brindley, 1955.

* Only appears after heating to 300° C. or above.

of both normal and heat-treated stevensite. This would suggest that the transformation of pectolite to stevensite took place above normal temperatures, probably before the final cooling of the sill.

There is close similarity between the North Tyne material and a mineral in the A.S.T.M. index reported as sepiolite. This mineral occurs as a coating on apophyllite that rests on pectolite from the Whin Sill at Copt Hill, and is almost certainly a type I stevensite.

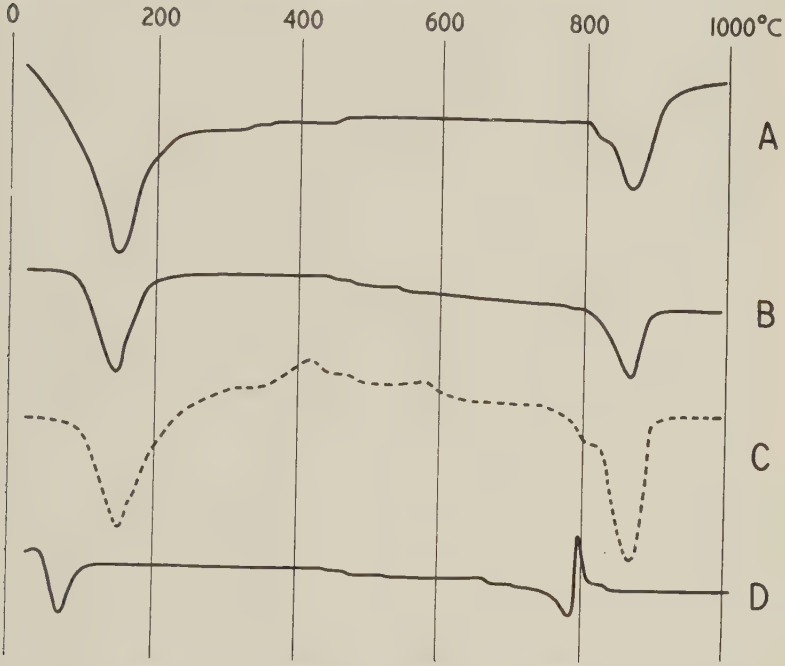


Fig. 2. Differential thermal curves of: (A and B) stevensite, type I, N. Tyne; (C) stevensite, Springfield, New Jersey (after Faust and Murata, 1953); (D) fibrous sepiolite, Ampandrandava, Madagascar (B.M. 1940, 40).

Differential thermal analysis. Fig. 2 shows D.T.A. curves for the North Tyne type I stevensite and a fibrous sepiolite from Ampandrandava, Madagascar, derived from analyses carried out on the same apparatus.[1] A curve for stevensite after Faust and Murata, 1953, is

[1] The thermal curves reproduced here were obtained with a medium sensitivity apparatus, without a pre-amplifier, using a 'Kent' potentiometer-type temperature-difference recorder and chromel-alumel thermocouples. The test sample was very small (less than 0·2 g.) and was sandwiched between calcined kaolin in a silica crucible. The thermocouples were placed horizontally and the crucible was finally tamped down with calcined kaolin.

also depicted as a dashed line; in this representation the various peaks are accurately positioned with regard to temperature, but the amplitude of the curve is not to the same scale as the others.

The curves for the North Tyne material compare favourably with the standard stevensite, showing two important endothermic peaks at about 150° C. and 860° C. and a small endothermic peak about 820° C. The sepiolite curve has two endothermic peaks at about 55° C. and 760° C. and a well-defined exothermic peak at about 780° C. Thus there is little similarity between the D.T.A. curves of the two minerals, and D.T.A. appears to be a satisfactory method of distinguishing between them.

It is interesting to note that the peaks on the curve for sepiolite occur at lower temperatures than those illustrated by Caillière and Hénin (1957). This variation is almost certainly due to the different conditions under which the analyses were carried out. The curves illustrated by Caillière and Hénin were obtained using an uncovered crucible (oxidizing conditions), while the new curves result from a study in a sealed crucible (reducing conditions).

Discussion and conclusions. The type I stevensite from the North Tyne region is an alteration product of pectolite, but the type II material may well be an original mineral in the amygdale-like structures found in the Whin Sill. Glenn (1916) suggests that stevensite from Springfield, New Jersey, was formed from pectolite. In the process he envisages, percolating rainwater becomes enriched in magnesia while passing through basalt, and the solution so formed reacts with pectolite transforming it into stevensite. If the strong line at 9·78 Å. is evidence of natural heat treatment, the above process could not apply in this case and the replacement of pectolite by stevensite would be penecontemporaneous with the infilling of the amygdales. The reacting solutions would then be magmatic, or partly so, at a temperature of about 400° C. It would appear, therefore, that Ca (to form pectolite) and Mg (to form stevensite) were concentrated in the residual phases of the Whin Sill magma of this area.

Many of the properties of stevensite closely resemble those of sepiolite, but D.T.A. is a satisfactory method of distinguishing between these two minerals.

Acknowledgements. I would like to thank Dr. R. A. Chalmers for the analysis of the stevensite, Dr. G. A. L. Johnson for the thermal analyses of both the North Tyne stevensite and the sepiolite, and Mr. J. Lee for draughtsmanship on the illustrations. I am also indebted to the Keeper of Minerals, British Museum (Natural History), for the sample of sepiolite from Madagascar used for D.T.A.

References.

Brindley (G. W.), 1955. Amer. Min., vol. 40, p. 239 [M.A. **13**–263].
Caillière (S.), 1936. Bull. Soc. franç. Min., vol. 59, p. 353 [M.A. **8**–225].
—— and Hénin (S.), 1957. The differential thermal investigation of clays, ed. R. C. Mackenzie, ch. 9, p. 231. London, Min. Soc.
Faust (G. T.), 1940. Rept. Invest. U.S. Dept. Inter., Bur. of Mines, R.I. 3522.
—— and Murata (K. J.), 1954. Amer. Min., vol. 38, p. 973 [M.A. **12**–494].
Glenn (M. L.), 1916. Amer. Min., vol. 1, p. 44.
Hendricks (S. B.) and Alexander (L. T.), 1940. Journ. Amer. Soc. Agronomy, vol. 32, p. 455.
Kauffman (A. J.), 1943. Amer. Min., vol. 28, p. 512 [M.A. **9**–123].
Longchambon (H.), 1937. Bull. Soc. franç. Min., vol. 60, p. 232.
Mielenz (R. C.), King (M.E.), and Schieltz (M. C.), 1950. Rept 7, Amer. Petroleum Project 49, Columbia University, New York.
Migeon (M. G.), 1936. Bull. Soc. franç. Min., vol. 59, p. 6 [M.A. **6**-477].
Ross (C. S.) and Hendricks (S. B.), 1945. U.S. Geol. Surv. Prof. Paper. 205B, p. 23 [M.A. **10**–26].
Shapiro (L.) and Brannock (W. W.), 1956. U.S. Geol. Surv. Bull. 1036-C.
Tomkeieff (S. I.), 1929. Min. Mag., vol. 22, p. 100.
Winchell (A. N.), 1951. Elements of Optical Mineralogy. 4th edn, part 2. New York (J. Wiley and Sons Ltd.).

Order and disorder in the feldspars, I.

By Helen D. Megaw, M.A., Ph.D., F.Inst.P.

Crystallographic Laboratory, Cavendish Laboratory, Cambridge, England.

[Read 4 June 1959.]

Summary. Feldspar structures are classified into small-cell ($c \approx 7$ Å.) and large-cell ($c \approx 14$ Å.) types, as a preliminary to a discussion of the nature of substitution disorder, position disorder, and stacking disorder, and of the relations between them. Diffraction effects characteristic of the different types of disorder are considered and related to experimental observation as far as possible.

THE feldspars comprise a large and extensively-studied group of materials of formula AB_4O_8, where A is a large, low-valent cation (K, Na, Ca, or Ba) and B includes both Si and Al. Between the ideal compositions, in which only one kind of A cation is present, there are large ranges of solid solution, and where these involve A atoms of different valencies there is a corresponding change in the Si:Al ratio to maintain electrical neutrality. An additional complexity consists in the fact that at most compositions 'high' and 'low' forms can be distinguished, which are believed to represent the quenched and annealed conditions respectively.

Much work has been done, by various techniques, to study the relationships between different members of the series, and in particular the nature and extent of the 'ordering' of Si and Al. Though the problem is physically quite different from that of ordering in alloys, the latter has been studied in so much more detail that it has affected much of the thinking about silicates and led to some confusion in terminology. Now that several structure studies on selected feldspars have replaced the vaguer ideas by more exact knowledge, it seems worth while to analyse the problem afresh and clarify our ideas of what we mean by order and disorder.

A list of the feldspars discussed in this paper is given in table I, together with references to recent detailed structure investigations. Names are to be taken, unless otherwise specified, as referring to the pure stoichiometric compound of ideal composition; and where the same name is commonly used for both high and low forms, the low form is meant unless the context makes it clear to the contrary.

TABLE I.

Mineral name	Chemical formula	High or low	Symmetry	Lattice	Cell dimensions (approx.) Å.	References to structure investigation
Microcline	$KAlSi_3O_8$	Low	Triclinic	*C*	8×13×7	Bailey and Taylor (1955)
Orthoclase	$KAlSi_3O_8$	Low	Monoclinic	*C*	8×13×7	Cambridge workers, unpubl.
Sanidine	$KAlSi_3O_8$	High	Monoclinic	*C*	8×13×7	Cole, Sörum, and Kennard (1949)
Albite	$NaAlSi_3O_8$	Low	Triclinic	*C*	8×13×7	Ferguson, Traill, and Taylor (1958)
	$NaAlSi_3O_8$	High	Triclinic	*C*	8×13×7	Ferguson, Traill, and Taylor (1958)
Anorthite	$CaAl_2Si_2O_8$	Low	Triclinic	*P*	8×13×14	Kempster, Radoslovich, and Megaw, unpubl.
	$CaAl_2Si_2O_8$	High	Triclinic	*I*	8×13×14	(Not obtainable in fully quenched condition)
Celsian	$BaAl_2Si_2O_8$		Monoclinic	*I*	8×13×14	Newnham and Megaw (1960)

Unit cells. The known feldspar structures are all built from variants of the same fundamental unit, a parallelepiped of dimensions approximately $a = 8{\cdot}6$ Å., $b = 13{\cdot}0$ Å., $c = 7{\cdot}0$ Å., $\alpha = 90°$, $\beta = 116°$, $\gamma = 90°$. Fig. 1 is a perspective diagram of the parallelepiped, and fig. 2 a schematic projection of the atomic arrangement. It forms a three-dimensional *B*-O framework in which every *B* atom is surrounded tetrahedrally by four oxygens, and every O is linked (not collinearly) to two *B* atoms; the *A* atoms occupy large cavities in this framework in which they may have from six to nine oxygen neighbours. The *B* sites are of two kinds, B_1 and B_2, distinguished electrostatically by the fact that the B_1 are linked to the unique O that has two *A* neighbours, the rest having one only.

There are differences from one feldspar to another not only in the dimensions of the parallelepiped and the chemical nature of the atoms occupying particular sites, but also in the detailed coordinates of the atomic sites. Further, they differ in the number of such parallelepipeds needed to constitute a true repeat unit. In some feldspars (e.g. albite) the parallelepiped is itself the unit cell, while in others (e.g. celsian) the true unit cell requires two such units, with identical dimensions but slightly different internal arrangements, sharing a common face parallel to (001), and thus giving rise to a doubled *c*-axis. This division into small-cell and large-cell feldspars, or 7 Å. and 14 Å. feldspars, is essential for our discussion. The fact that in some (e.g. sanidine) there is a mirror plane of symmetry within the unit cell, while in others (e.g. albite) there is only a rough approximation to one, is of secondary importance.

In the simpler feldspars, the parallelepiped is face-centred on *C*, so

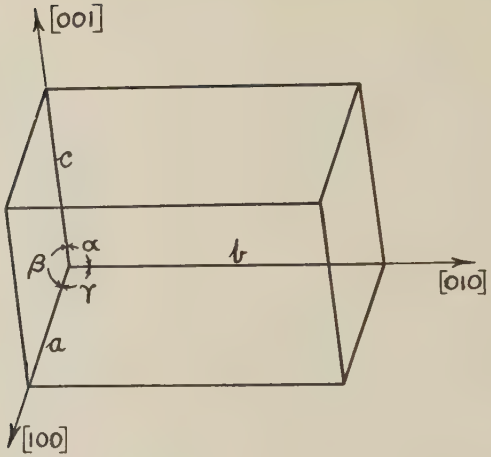


Fig. 1. Unit cell of 7 Å. feldspar, e.g. albite, on conventional axes.

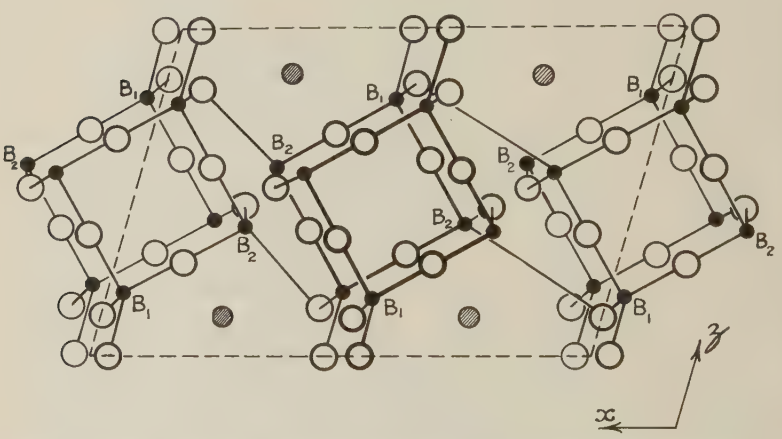


Fig. 2. Stylized diagram of feldspar structure, e.g. albite, projected on (010), to show linkages within the (Si, Al)–O framework. Pairs of atoms related by a pseudo mirror plane have arbitrarily been given a constant relative displacement; linkages to atoms above and below the unit cell shown have been omitted. Small filled circles, Si or Al; large open circles, O; hatched circles, cation *A*.

that a smaller primitive unit cell could have been chosen in various ways, most simply by taking $\frac{1}{2}(\mathbf{a}+\mathbf{b})$ instead of $\mathbf{b}$ for a cell edge. This is shown in fig. 3 by the parallelogram *ORST*, where *OPQR* represents the base of the conventional *C*-centred cell. In anorthite the face-centring has disappeared, and the cell with base *OPQR* is primitive; but we could

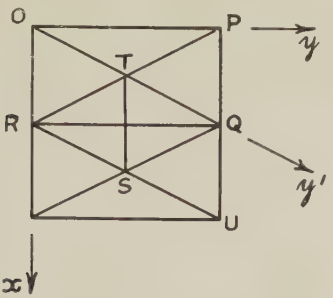


FIG. 3. Base of unit cell of feldspar in (001)-plane. *OPQR*, conventional unit cell of albite (*C*-centred) and anorthite (primitive). *ORST*, primitive unit cell of albite. *OQUR*, alternative choice of primitive unit cell for anorthite.

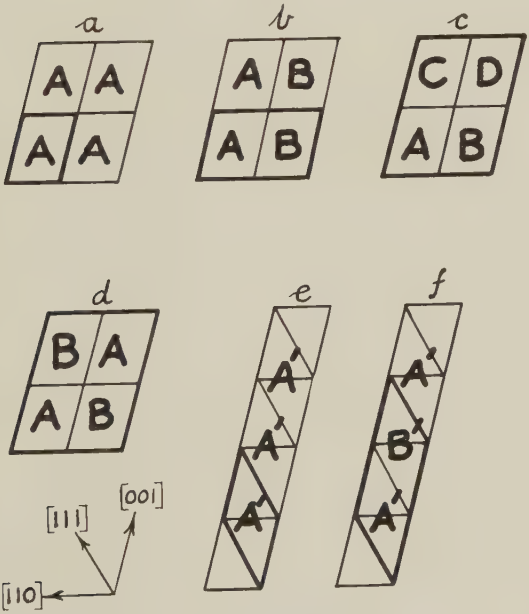


FIG. 4. Relations between true cells and albitoid sub-units for several feldspars: *a*, albite; *b*, hypothetical 7 Å. primitive cell; *c*, anorthite; *d*, celsian; *e*, albite (alternative choice of primitive cell); *f*, celsian (primitive cell). Directions in key are indexed using conventional albite axes.

equally well have chosen the cell with base *OQUR*, which is convenient because its axes are those of a primitive albite cell. Fig. 4 shows comparisons between several feldspar structures in the plane of *OQ* and the

c-axis ($\bar{1}10$) with conventional albite indexing). Fig. 4*a* shows four primitive albite cells, all identical. Fig. 4*b* is a hypothetical structure, of which no actual example is known, where the face-centring has been lost without any change in the c-repeat length; the true cell contains two albitoid sub-cells. Fig. 4*c* shows a primitive cell of anorthite, consisting of four non-identical albitoid sub-cells.

Celsian, when described on conventional axes, has the same cell dimensions as anorthite but is body-centred (Newnham and Megaw, 1960). If we use the primitive-albite axial directions, it can be seen from fig. 3 that it must be described as A-face-centred, and this description allows the comparison with anorthite shown in fig. 4*d*; the unit cell contains four albitoid sub-cells consisting of two identical pairs. If we wish to work with a primitive repeat unit (which is here less convenient) it is most easily described by reference to a differently-chosen primitive albite cell, shown in fig. 4*e*, whose b axis lies along the [111] conventional albite direction. From fig. 4*f* it can be seen that the primitive celsian cell contains two non-identical sub-cells.

Effects in large-cell structures. If the sub-cell instead of the true cell were used to define a lattice for one of these large-cell structures, it would appear that the coordinates of all atoms varied systematically so as to give displacements from a mean position that had equal magnitudes but opposite signs in alternate sub-cells. This description in terms of displacements from mean positions is of course fictitious, though it may be a useful fiction. To understand the effect in physical rather than geometrical terms, it is necessary to realize that the alternation of sub-units is the equilibrium configuration of the whole set of atoms. The stable arrangement does not quite fit a repeat unit with the dimensions of the sub-cell, but the misfit can be compensated when two slightly different arrangements alternate to give the true cell.

Large-cell structures give diffraction maxima at positions determined by the true repeat, but the intensities of these, except when they coincide with small-cell maxima, are due to differences in the positions of the atoms in the sub-cells, and to differences in scattering factor if the chemical contents are not identical; since the differences are, by hypothesis, small, the intensities are small. These *difference reflections* (more commonly, but confusingly, known as 'superlattice reflections') are characteristic of the large cell. In anorthite-like feldspars they have been classified and named 'b', 'c', and 'd' types, the reflections at positions common to the albite and anorthite cells being 'a' (Gay, 1953; Laves and Goldsmith, 1954).

If in any structure investigation the difference reflections are overlooked altogether (as may happen because of their weakness) the resultant electron-density map is an average of those of the sub-cells, and suggests a scatter of the atomic positions about a mean, and hence disorder. If the difference reflections are included with their correct sign, or phase, the true structure is displayed. A deliberate omission of the difference reflections for obtaining an average map has been a very useful preliminary step in several structure analyses of feldspars.

It should be emphasized that large-cell structures are in themselves no indication of disorder; indeed, their occurrence suggests rather that the need for disorder has been eliminated. They warn us, however, that we may need to examine the related small-cell structure for the presence of disorder.

Types of disorder.

We define a perfectly-ordered structure, or perfect structure, as one built up by translation-repeat of parallelepipeds identical in size, chemical content, and positions of atoms within the parallelepiped, except for the effect of thermal vibrations. We idealize the situation by neglecting all impurities and mechanical defects.

Three types of disorder may be distinguished: substitution disorder, position disorder, and stacking disorder. Other types that have been recognized in other materials, such as vacant sites or interstitial atoms, are not relevant here; nor are the more complicated effects involving symmetry operations other than translation (the '*OD*' effects described by Dornberger-Schiff, 1956). For the purpose of the present discussion, which is concerned chiefly with the effects of the three types of disorder on the X-ray structure determination, we need make no assumptions as to the mechanism of the processes responsible for the occurrence of disorder; some aspects of these processes will be considered elsewhere in relation to growth and the subsequent thermal history of the feldspar.

Substitution disorder.

This occurs when one crystallographically-equivalent set of sites carries atoms of more than one chemical species distributed at random over the set of sites. It is characteristic of solid solutions (defined as materials in which individual atoms of the 'solvent' or host structure are replaced over a continuous composition range by chemically similar atoms of the 'solute' or guest); but it can also occur in stoichiometric materials containing two very similar atoms, where their disordered

distribution between the available sites constitutes a sort of internal solid solution.

It is important here to distinguish between sites which are crystallographically equivalent and those which are merely chemically similar, in the sense that they provide similar environments for the atoms in them. While crystallographic equivalence is an absolute, chemical similarity may show degrees of closeness. Thus, for example, all the B sites in any feldspar are chemically similar to a first approximation, yet there are, as we have seen, differences between B_1 and B_2 which may be important when one comes down to details. It is not often possible to predict whether the chemical similarity between two given sets of sites is sufficient to ensure that they will be equally suitable for guest atoms. In anorthite, for example, there are four independent sets of A sites, and we cannot say *a priori* whether Na will replace Ca in one set only, or in all sets with equal probability, or with a different probability for each set.

When the guest atom differs in size from the host, the coordinates of its nearest neighbours are affected; but if these can adjust themselves easily with respect to their other neighbours the disturbance remains local, and unless the guest atoms are very numerous there is little tendency to order. The consequent random variations of coordinates from one unit cell to the next give rise, on electron-density maps, to a spreading-out of the atoms like that produced by thermal vibrations. If the effect is small, as it usually is, it cannot be distinguished from that of an 'average atom', or mean of the host and guest atoms weighted according to their probability of occurrence, with corresponding average positions for the neighbouring atoms. As far as the intensities of X-ray diffraction maxima are concerned, the structure is a *quasi-perfect* structure of average atoms.

This gives us our standard method of studying Si–Al substitution disorder in the feldspars, a method first used for orthoclase by Chao, Hargreaves, and Taylor (1940). It is assumed that the distance B–O for an average atom varies linearly with its composition. Then exact determination of the B–O distance for each set of sites gives us the composition of the average atom occupying this set.

Evidence from structure determination. The results of structure determination show that in *all* the low feldspars studied (orthoclase, microcline, albite, anorthite, celsian) there is an approach to an idealized perfectly ordered arrangement, except for orthoclase, where there is some evidence (now under re-examination) that while the occupation of

the B_2 sites approaches complete order, that of the B_1 sites is random. It is not yet certain whether the residual disorder observed experimentally in some cases is within the limits of experimental error, or is an effect of imperfect annealing, or persists even in the best material; in any event it is not so large as to obscure the idealized picture, and it seems most likely that an understanding of the system as a whole can best be achieved by taking the perfectly ordered structures as starting-points for discussion. In this paper we shall not attempt to go beyond this idealization to the second approximation, which considers residual disorder.

For the high temperature structures (sanidine, high albite) there is complete disorder, with equal distribution on all sites. It is to be noted that this change from order in the low-temperature form to disorder in the high-temperature form (experimentally irreversible under laboratory conditions at present attained) is in accordance with the evidence from thermal studies, from which in fact it was predicted.

The statement that there is complete order, in the crystallographic sense, does not tell us the whole story; we want to know the relative arrangement of Si and Al in the ordered distribution. (In the literature, the two ideas have sometimes been run together confusingly.) To describe this, we notice that the two independent sets of sites in orthoclase and sanidine, B_1 and B_2, are subdivided into four sets in albite and microcline, $B_1(0)$, $B_1(m)$, $B_2(0)$, $B_2(m)$ (Si_1, Si_1', Si_2, Si_2' in the old notation): and each of these is again subdivided into four sets in anorthite; in the 14 Å. celsian cell the two sets B_1 and B_2 are divided into four in a different way, conforming to body-centred monoclinic symmetry. In albite and microcline, $B_1(0)$ is occupied by Al, the other sites by Si. In anorthite, however, half the four sets derived from *each* of the original four, $B_1(0)$, $B_1(m)$, $B_2(0)$, $B_2(m)$, are occupied by Al and half by Si, and exactly the same distribution is found in celsian, in spite of its different symmetry. This distribution (which is somewhat idealized, since perfection of order is not observed in the actual materials) is characterized by a perfect alternation of Si and Al along any line of B–O bonds. It incorporates the 'aluminium-avoidance' rule suggested by Loewenstein (1954). It can be predicted on electrostatic grounds, as it tends to equalize the electrostatic valence bonds to all oxygens. (See also Goldsmith and Laves, 1955, and De Vore, 1956.) It is satisfactory that it has now been confirmed for anorthite and celsian by direct structure determination. We may note that both the 14 Å. repeat period and the 1:1 ratio of Si:Al are necessary to allow this arrangement.

In solid solutions, of course, perfect order cannot be retained either for the A or for the B sites. If we start from the nearest perfect structure, we expect to find that some but not necessarily all of the sets of sites have become randomly occupied. Little information is yet available experimentally, but there is one known feature of the Si–Al arrangement in the plagioclases that may also be of considerable importance in other feldspars: the arrangement in albite cannot be derived from that of anorthite by continuous replacement of Al by Si, but requires also the replacement of Si on half the $B_1(0)$ sites by Al. This is likely to be a more difficult reorganization, and its onset should therefore be recognizable as corresponding to some break in the continuity of the series. It seems possible that steady replacement of Al by Si on B_2 sites may take place from pure anorthite to about $An_{25}Ab_{75}$, and that the peristeritic effects which then set in, with the formation of two phases, may represent the different kinds of Si–Al arrangements needed on the B_1 sites to secure electrostatic balance at this composition. The orthoclase–microcline relationship may also involve a reorganization of this kind.

Position disorder.

It has been suggested that in the feldspars the A cation may have position disorder. The assumption is that, while the B–O framework repeats perfectly with the periodicity of the small cell, the cavity formed by it allows two (or more) sites for A so nearly equal in energy that they are occupied at random. If their separation in space is small, the electron-density map will look as if A had a large anisotropic temperature factor; if the separation is large, the single elongated peak may be resolved into two half-atoms. As with substitution disorder, the structure is quasi-perfect, but this time it incorporates half-atoms instead of average atoms.

Experimentally, an effect like this has been observed for Na in high-albite by Ferguson, Traill, and Taylor (1958), who suggest that it may represent either position disorder or a true anisotropic temperature factor. A third possible explanation will be mentioned later (p. 239).

Position disorder has also been invoked (Laves and Goldsmith, 1954, and Goldsmith and Laves, 1955) to explain the low–high transition in anorthite, which involves the reversible disappearance or change of sharpness of certain difference reflections, the c and d types, corresponding to a change of the unit cell from primitive to body-centred. This explanation assumes that the B–O framework is nearly body-centred at all temperatures, and that it contributes negligibly to these reflections,

which are in effect entirely due to Ca, ordered on different sites within the cavities in the low form, disordered between the same sites in the high. From our knowledge of the structure of low anorthite (Kempster, 1957) we can now tell that this explanation is neither necessary nor sufficient. The *B*–O framework is not very close to body-centred, but forms four independent pairs of cavities each occupied at a specified position by Ca: its contributions to the *c* reflections are not negligible. The transition to a body-centred form necessarily involves a straightening out of the framework, qualitatively of the same kind as occurs in transitions in the perovskite family. If the framework becomes body-centred the set of sites available for Ca becomes body-centred also, and there is no need to postulate disorder while the material remains at the high temperature. When it is quenched, stacking disorder may be introduced; this will be discussed below.

Stacking disorder.

In this we are no longer dealing with individual atoms but with pseudo-translation-repeats or sub-units. Disorder of this kind consists in a break in the ordered sequence of sub-units. We may illustrate the important features with a two-dimensional example where the true unit cell consists of two sub-units *P* and *Q*. Two kinds of wrong sequence are possible: in a row of cells in the [001] direction (fig. 5*a*) we may have two consecutive *Q*'s, e.g. ...*PQPQQPQP*... or in a row in the [010] direction (fig. 5*b*) we may change from *P* to *Q*, e.g. ...*PPPPQQQ*.... If either kind of mistake happens in the same cells in adjacent rows, we get boundary planes parallel to (001) and (010) respectively, separating two regions of perfect crystal. We call these boundaries *fault planes*.

If we try to introduce either kind of mistake at random, not building up into fault planes, as in fig. 5*c*, we find we have introduced the other as well. An arrangement such as this looks as if it would be smoothed out by annealing. If *P* and *Q* differ only in their atomic coordinates and not in their chemical contents -as happens in the rather more complicated example of anorthite, for the sub-cells related by a body-centring translation—a perfect structure should fairly easily be achieved, since the shift needed for any individual atom is small. This may very well be the explanation of the high low transition in anorthite. But if the sub-cells differ chemically in their contents, the process is more difficult. What happens is that, by diffusion of atoms, the boundaries move until a position of minimum energy is attained. Whether this is like fig. 5*a* or 5*b*, with one type of mistake and one type of fault plane only, like fig. 5*d*

with a single stepped boundary in a definite crystallographic direction, or like fig. 5*e* with intersecting fault planes, will depend on the particular nature of the material and perhaps on its thermal history.

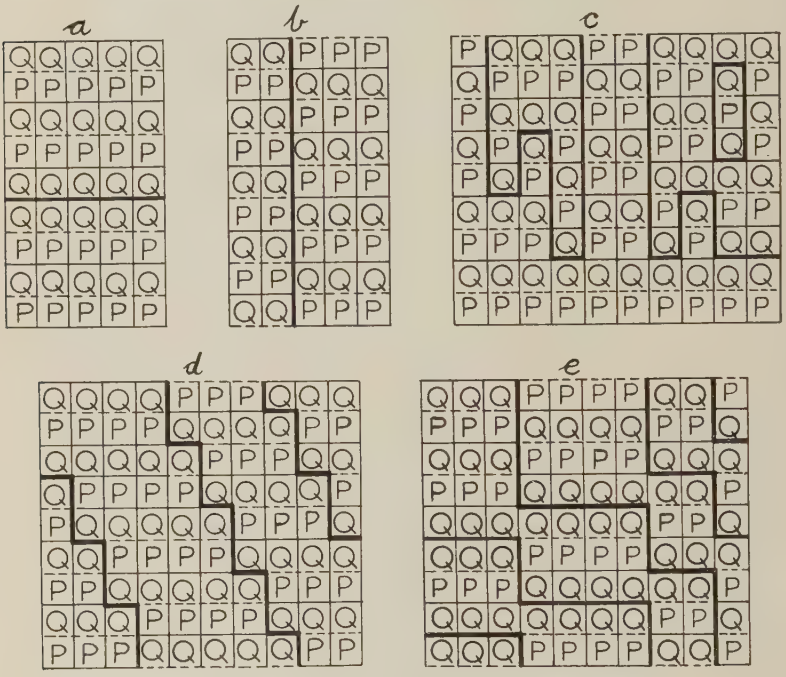


FIG. 5. Different distributions of mistakes in a hypothetical structure *PQ*: *a* and *b*, single fault planes in different directions; *c*, random distribution of mistakes; *d*, combination of two types of fault planes to give effective fault planes parallel to a third direction; *e*, independent occurrence of two types of fault plane.

The hypothetical example in fig. 5 is formally like CuAu. Other examples with more than two sub-cells are reminiscent of the 'anti-phase domains' in copper-gold alloys (see, for example, Lipson, 1950), and in fact the same principle underlies both; but the theories of anti-phase domains have been worked out there for the particular case when the sub-cell corresponds to the primitive unit cell of a face-centred cubic lattice, and contains only one atom.

The X-ray diffraction effects from a structure containing stacking faults are no longer those of a perfect or quasi-perfect grating. For the perfect structure whose repeat unit consists of two sub-units, we have seen that the pattern is made up of small-cell reflections and difference

reflections. For the faulted structures of fig. 5, if we consider diffracted radiation in the directions which, for a perfect structure, would be first-order maxima, we see that phase differences of π are introduced at the faults. If the faults are numerous, these maxima are wiped out. The same is true for *all* odd-order maxima, i.e. for all the difference reflections. An electron-density map constructed from all the remaining maxima will show the average of sub-cells P and Q, as in the case of substitution disorder, but now, instead of spread-out atoms in the neighbourhood of the average atom only, we expect to find every atomic peak doubled. The distinction between the two is likely to be less marked in practice than in theory, because small separations are hard to detect with certainty.

This, however, is not the whole story. If the faults are in any way systematic, the intensity in directions other than the maxima may build up significantly, and give rise to measurable reflections. (The quasi-perfect structures of course give a background intensity different from that of the perfect structures, but the differences are very much smaller than those with which we are here concerned.) The extra reflections that occur in the intermediate plagioclases, from about $Ab_{25}An_{75}$ to $Ab_{75}An_{25}$, are believed to be due to this effect. They will be discussed more fully elsewhere.

Discussion.

Relations between the three types of disorder.

While the three types can be discussed separately, they are not in fact independent. We assumed, in dealing with substitution disorder, that effects of a guest atom ceased to be felt beyond its near neighbours. This is unlikely to be true, especially if, as in the plagioclase feldspars, the guest atom has a different electrostatic charge from the host. The most important effect of an Na–Ca substitution, for example, is probably not so much on the positions of its nearest O neighbours as on the Si–Al distribution among its second-nearest neighbours, and through them out to distances of the order of magnitude of the sub-cell. In other words, substitution on a particular site tends to produce regions of local order different from the order of the host structure.

Beginning at the other extreme, we notice that changes in the P–Q sequence are likely to be brought about by substitution if the structural difference between the sub-cells is such that one sub-cell can accommodate the guest atom more easily than the other. This suggests that

a fault has its origin in a site mistake. If we picture it as arising during growth, and suppose that sub-cell Q can accommodate Na more readily than P can, then when Na is deposited on what should be a Ca site it will affect its surroundings so that Q is built instead of P, giving rise to a fault. During subsequent annealing, Na (and perhaps Si with it) diffuses through the structure, and the fault travels along a row of cells, rather as dislocations do, until it combines with faults in other rows to form a fault plane.

This example is only put forward as a hypothetical one to illustrate the possibilities. The models required to explain actual effects in feldspars have a more complicated set of sub-cells and fault planes.

Probability of site mistakes.

The probability of a site mistake will in general depend on the number of guest atoms available, and on the environment of the site concerned. To obtain a picture of the sort of thing that is happening, it is worth distinguishing certain extreme cases.

In the first, the *simple site mistake*, the probability of a mistake on any permissible site is the same; it is independent of the existence of other mistakes, and is equal to the abundance ratio of the guest atom, i.e. the ratio of the number of guest atoms to the total number of atoms able to enter the site concerned, after atoms required for fully ordered sites have been subtracted. This will be a linear function of the composition.

At the other extreme is the *interchange site mistake*. In this there is a pair of sites such that the guest atom can enter the one normally reserved for the host provided that at the same time the host atom enters the one normally reserved for the guest. The probability of this kind of site mistake is independent of composition, except through the indirect effect of neighbouring mistakes.

We may also consider the possibility of *paired site mistakes*. These will occur if the sites are grouped in pairs so that a mistake must occur on both if it occurs on either; if all sites are in pairs, the probability is exactly the same as for a simple site mistake.

This list is not meant to be exhaustive, and is put forward merely to clarify the possibilities: it is not known yet what actual examples occur.

Probability of faults.

If faults have their origin in site mistakes, the probability of occurrence of a fault is linked through them with the composition. It may be that

one site mistake on a particular site is sufficient to nucleate a fault; it may be that several specified site mistakes are required; on many sites, perhaps on most sites, mistakes will be ineffective. In general, the probability of a fault is thus some function of the composition.

The 'fault' boundary is in fact a region of perfect two-dimensional order (except where it is intersected by other faults), of thickness about one unit cell. Each half is continuous with the perfect structure on that side of it, but the two halves together constitute something new, and have a different chemical composition from the rest. When a nucleus is laid down, annealing gives rise to lateral growth, by diffusion of ions from the immediate environment to sites of lower energy in the boundary structure.

Character of disorder in feldspars.

We must return to the question whether, in the high-temperature feldspars, the occupation of B_1 and B_2 sites by Al and Si is completely random, or whether there is local order based on a large unit cell with faults. It seems very improbable that there should be no sort of local order. On the other hand, if the faults exist they are not organized so as to give extra maxima, and hence the only information available to us is the electron-density map of the average sub-cell. If, on this, it were possible to recognize each peak as the combination of some small number of nearly coincident peaks—say two or four—and if these could be sorted out so as to give two or four B-O networks each with reasonable interatomic distances, it would be a strong argument in favour of the large-cell description with stacking faults. (The problem of sorting out the set of multiple peaks into separate networks is like that solved by Carlisle and Crowfoot (1945) for more widely separated peaks in cholesteryl iodide.) Practically, however, it is unlikely that a structure analysis of sufficiently high resolution could be achieved. The most that might be expected is that a few multiple peaks could be detected which were consistent with such an hypothesis.

The limited amount of information available rather favours the hypothesis. It was mentioned above, p. 234, that high albite has an elongated peak for Na, which may be interpreted as due to anisotropic temperature factor or to position disorder. It may equally well mean that the observed unit cell is an average of two sub-units of a large cell in which only Na has conspicuously different coordinates. If the more detailed examination now in progress should reveal separation of peaks of neighbouring atoms it would be evidence for this point of view.

The difference between orthoclase and microcline may be discussed in terms of stacking disorder. As a first approximation, the Al atoms in a maximum microcline may be supposed completely ordered in the $B_1(0)$ sites of a 7 Å. cell, while those in orthoclase appear to be distributed at random between the $B_1(0)$ and $B_1(m)$ sites (which are indistinguishable in monoclinic symmetry) of the same size of cell. Suppose, however, that we had a 14 Å. structure in which Al was ordered on half the B_1 sites in the same way as in anorthite, while all B_2 sites were occupied by Si, and suppose that this structure had a random set of faults on planes with zone axis [001], whose slip vector had a component $c/2$: this would be indistinguishable from orthoclase, within the limits of accuracy so far achieved. It is an essentially different picture from that of submicroscopic twinning put forward (Laves, 1950; Goldsmith and Laves, 1954) to explain orthoclase. A refinement of the orthoclase structure, now in progress in this laboratory, may perhaps throw light on this question.

Conclusion.

This paper attempts to set out the important physical features of a complex problem in which much confusion has arisen, partly from a tendency to assume that concepts developed for application to order–disorder phenomena in alloys can be used fairly directly in relation to effects observed in feldspars. As far as possible the discussion is based on factual information, much of it derived from recent accurate analyses of type structures in the feldspar group; where speculative suggestions are advanced, they are clearly distinguished as such, and their eventual confirmation or disproof will leave the main account unaffected.

It is worth while to point out that although the present treatment is confined to the feldspars, a more detailed development of some of the fundamental ideas, to be published elsewhere, makes it clear that they may be of considerable importance in their application to other minerals.

Acknowledgements. I wish to express my indebtedness to Dr. W. H. Taylor for his encouragement and help throughout the investigations of which this forms a part. I am grateful to McMaster University, Hamilton, Ontario, for hospitality during the time while this paper was being written. Many of the ideas discussed in the paper have arisen in connexion with the development of structural researches recently completed in the Cavendish Laboratory; we are indebted to the Nuffield Foundation for the award of generous grants in support of this programme.

References.

Bailey (S. W.) and Taylor (W. H.), 1955. Acta Cryst. vol. 8, p. 621.
Carlisle (C. H.) and Crowfoot (D.), 1945. Proc. Roy. Soc., ser. A., vol. 184, p. 64.
Chao (S. H.), Hargreaves (A.), and Taylor (W. H.), 1940. Min. Mag., vol. 25, p. 498.
Cole (W. F.), Sörum (H.), and Kennard (O.), 1949. Acta Cryst., vol. 2, p. 280.
De Vore (G. W.), 1956. Zeits. Krist., vol. 107, p. 247.
Dornberger-Schiff (K.), 1956. Acta Cryst., vol. 9, p. 593.
Ferguson (R. B.), Traill (R. J.), and Taylor (W. H.), 1958. Ibid., vol. 11, p. 331.
Gay (P.), 1953. Min. Mag., vol. 30, p. 222.
Goldsmith (J. R.) and Laves (F.), 1954. Geochimica Acta, vol. 6, p. 100.
—— 1955. Zeits. Krist., vol. 106, p. 213.
Kempster (C. J. E.), 1957. Thesis, Cambridge University.
Laves (F.), 1950. Journ. Geol., Chicago, vol. 58, p. 548.
—— and Goldsmith (J. R.), 1954. Acta Cryst., vol. 7, p. 465.
Lipson (H.), 1950. Progress in Metal Physics, vol. 2, p. 9.
Loewenstein (W.), 1954. Amer. Min., vol. 39, p. 92.
Newnham (R. E.) and Megaw (H. D.), 1960. Acta Cryst., in press.

A note on pyrosmalite from Nant Francon, North Wales.

By P. E. Brown, B.Sc., Ph.D., F.G.S.

Department of Geology, University of Sheffield.

[Taken as read 4 June 1959.]

Summary. The presence of pyrosmalite is recorded in a metasomatized slate that is in contact with intrusive rhyolite in the Nant Francon area of North Wales.

PYROSMALITE has been found in a metasomatized slate from Nant Francon. The rock containing the mineral was first described by Williams (1930) in a paper dealing with the geology of the country between Nant Peris and Nant Francon. In this paper Williams briefly described the metamorphic effect of the Bwlch-y-Cywion intrusion on the Ordovician slates which form the country rocks. Near the old Nant Francon road, north-west of Blaen-y-Nant, where the slate is in contact with intrusive rhyolite, there is a small exposure of what Williams called a biotite-garnet rock, developed from the normal chloritic slate of the locality by 'additive' metamorphism.

It has now been found that the minerals contained in the altered slate are: a green mineral of chlorite type, the exact identification of which is not yet certain, and also a brown oxidation product of this; almandine-spessartine garnet; iron ore; apatite; and pyrosmalite.

This is the first recorded occurrence of pyrosmalite in the British Isles. According to Frondel and Bauer (1953) pyrosmalite has been recorded only from the magnetite deposits of Nordmark and Dannemora in Sweden, and manganpyrosmalite from the franklinite deposit at Sterling Hill, New Jersey (Frondel and Bauer, 1953), and from Broken Hill, New South Wales (Hutton, 1956).

The normal unaltered slate in the vicinity of Blaen-y-Nant has well developed cleavage and is composed of chlorite with small grains of quartz, feldspar, and some magnetite. Against the rhyolite the slate has been altered to a massive uncleaved rock, very dark green in colour and weathering to a rusty brown. The complete transition to unaltered slate takes place in some 10 to 20 yards from the contact.

The metasomatized rock is variable. Some sections contain only rounded garnets in a groundmass of the green micaceous mineral, and minor iron ore; in other sections iron ore is a major constituent. Apatite (identified by means of an X-ray powder photograph) occurs sporadically as poikilitic plates up to 2 to 3 mm. across. Pyrosmalite is only a very minor constituent of the rock and occurs as clusters of radiating crystals

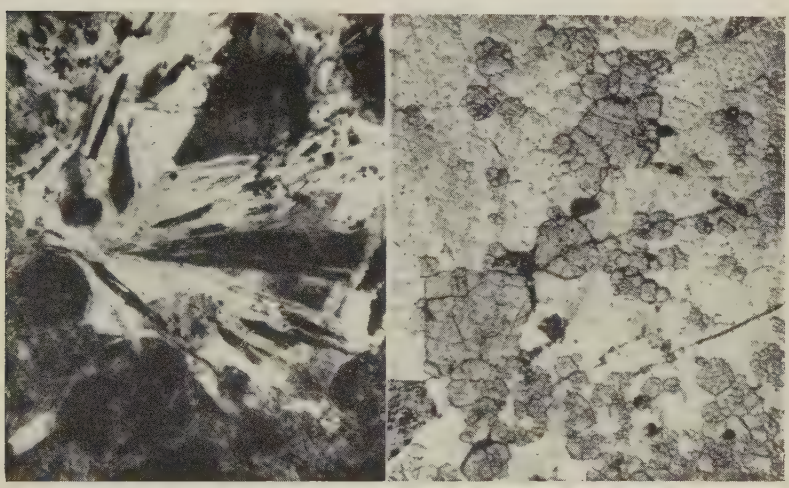


Fig. 1. Fig. 2.

Fig. 1. Pyrosmalite with garnet, chlorite, and iron ore in metasomatized slate from Nant Francon, North Wales. Crossed nicols. ×100 approx.

Fig. 2. Single crystal of apatite enclosing garnets in metasomatized slate, Nant Francon. Plain polarized light. ×100 approx.

of the order of 0·5 mm. in length. Some sections of the rock are traversed by veins, clearly visible in some hand specimens, between 0·5 and 1 mm. in width, and composed either of carbonate or of well-developed flakes of the green micaceous mineral. Preliminary chemical work on the unaltered slate and its metasomatized equivalent indicates that the major addition has been iron, with some manganese and smaller amounts of magnesium.

Attempts to separate the pyrosmalite by normal methods using heavy liquids and an electromagnetic separator were not successful, but sufficient of the mineral for an X-ray powder photograph was picked out of a thin section. The X-ray powder pattern agrees well with that of manganpyrosmalite (Frondel and Bauer, 1953). In view of the

preponderance of iron over manganese in the Nant Francon rock, it seems likely that the mineral is pyrosmalite rather than manganpyrosmalite. The mineral occurs as radiating clusters of grains with poor basal cleavage. It is uniaxial, negative, and the refractive indices determined on a grain picked out of a thin section are ϵ 1·638 and ω 1·662. The refractive indices of pyrosmalite (Winchell, 1951) are ϵ 1·647, ω 1·682, and those of manganpyrosmalite from New Jersey are ϵ 1·631, ω 1·669 (Frondel and Bauer, 1953).

A further account of the chemistry and mineralogy of this unusual rock will be prepared. Preliminary work has already been carried out, but owing to the fine grain size and abundance of iron ore inclusions, pure mineral fractions are difficult to obtain.

Acknowledgements. The work was carried out in the Department of Geology, Manchester University. The writer is indebted to Dr. G. D. Nicholls who first noted the unusual nature of the rock and suggested that its mineralogy would repay investigation, to Dr. J. Zussman for help with the X-ray data and for identifying the powder photograph as that of pyrosmalite.

References.

Frondel (C.) and Bauer (L. H.), 1953. Amer. Min., vol. 38, p. 755.
Hutton (O. C.), 1956. Ibid., vol. 41, p. 581.
Williams (D.), 1930. Quart. Journ. Geol. Soc., vol. 86, p. 191.
Winchell (A. N.), 1951. The elements of optical mineralogy, Pt. 2, 4th edn, New York (Wiley & Sons Ltd.).

Note. A coarsely granular specimen of greenish brown pyrosmalite, B.M. 1958,89, with individual cleavages up to 5 mm. across, has been presented to the British Museum (Natural History) by Mr. A. W. G. Kingsbury; it was collected by the donor at Treburland mine, Altarnun, Cornwall, and its identity has been checked by X-ray powder photograph.

P.G.E.

A new method for orthoscopic adjustment with the Fedorov stage.

By A. F. Hallimond, M.A., Sc.D.

75 Corringham Road, London, N.W. 11.

[Read 2 April 1959.]

Summary. A new method is described to replace the standard procedure for completing the adjustment of an optical symmetry plane, with conditions for the accurate use of the biquartz. Extinction measurements are plotted to show the wide range of sensitivity (for the adjustment of A_1 and A_2) at various directions in the symmetry planes of a plagioclase. It is proposed that A_1 and A_2 should be adjusted at different settings of A_4 so as to obtain maximum sensitivity for each of those axes.

THE standard procedure for adjusting a symmetry plane normal to the axis A_4 involves tilting about A_2 and A_4 and comparing the persistence of extinction. It has been described in nearly every textbook and is 'the most difficult and unsatisfactory feature of universal stage manipulation' (Emmons, 1931, p. 131).

For accurate work it is essential to realize that the sensitivity of adjustment is not only widely variable for different directions in the indicatrix, but that even for a given direction it may be completely different for A_1 and A_2. For example, when an optical symmetry axis is vertical in an adjusted symmetry plane, extinction persists for all values of A_2, which therefore cannot be given a precise setting, while A_5 and A_1 are very sensitive, since this direction is at the centre of the well-known conoscopic 'flash-figure'.

The method now proposed for completing the adjustment of the symmetry plane consists in examining the sensitivity for various settings of A_4. The setting of A_1 and A_2 is then completed by using those directions in A_4 along which the sensitivity has proved suitable.

Procedure. The microscope must be in complete adjustment. The condenser and any other substage optical fittings (except the weak auxiliary lens below the polarizer) must be removed. The condenser iris is retained as a screen to limit the beam to the useful part of the hemispheres, but need not be accurately centred. By means of the flat mirror the beam of the lamp is reflected axially so that an image of the lamp

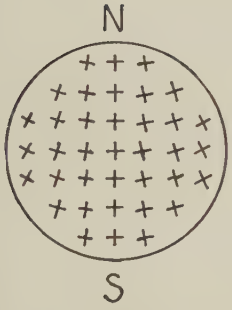
iris is seen at the back of the objective. This disc is carefully centred and the mirror is locked in position with the finger screws. The size of the disc is controlled by the lamp iris: it should not be as narrow as the objective iris opening, since the disc sometimes moves a little when the hemispheres are rotated. It may be twice the diameter chosen for the objective iris, or more. *It is essential that this disc should be completely and evenly illuminated*: a ground glass must therefore be inserted in the lamp after focusing, so as to break up any image of arc or filament.

The beam used for observation must be strictly coaxial with the microscope tube and of a conoscopic aperture (in the $5\times$ objective) about two degrees. This is approximately the diameter of the conoscopic reading mark in a Cooke objective (Manual, p. 143). Light outside this cone is excluded by the objective iris, which is contained in a section that can be screwed into the tube above the objective (if the objective does not already contain an iris). When the objective iris is nearly closed it has the same diameter as the reading mark; it then somewhat impairs the definition of the image, but this does not prevent accurate setting. During ordinary observation this iris must be opened; it should therefore have very easy movement (thick grease if present should be removed).

The biquartz. Although settings can be made by extinction, the biquartz has great advantages both in accuracy and in ease of adjustment. Tests with a Macé de Lépinay wedge showed that in the present measurements of a plagioclase a suitable rotation was $\pm 7°$. The Nakamura plate with only $1{\cdot}2°$ gives too little contrast under similar conditions. The biquartz plate is inserted in a slotted ocular which is preferably of the positive (Kellner) type in which no lenses are interposed between the analyser and the objective. The biquartz boundary must cross the centre of the field, since non-axial points are subject to small field errors.

With the universal stage the orthoscopic action of the biquartz differs from that in a homogeneous beam (e.g. in the ore-microscope). The light at any point in the field contains all the rotations belonging to rays coming from the sides of the isogyre, up to the boundary set by the objective iris, i.e. up to one degree or more from the trace of the optical symmetry plane (fig. 1). When the symmetry plane accurately bisects the iris opening, the effect upon the biquartz of light coming from the two sides will be equal and opposite and the biquartz will be matched. *It is essential, however, that the two sides of the isogyre shall be illuminated with equal intensity*: the back of the objective must show a uniformly illuminated disc bounded by the objective iris. It is often permissible

to open the iris to a somewhat wider diameter (say $\pm 2°$). There is no definite extinction position for either side of the biquartz, but a marked failure in matching follows on a small displacement of the optical symmetry plane.



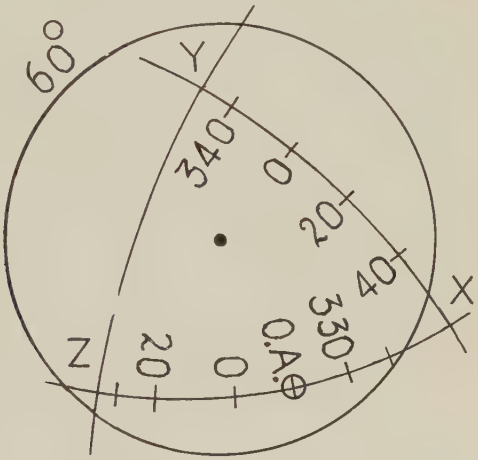


Fig. 1. Fig. 2.

Fig. 1. Vibration directions in the objective iris with an optical symmetry plane in adjustment.

Fig. 2. Stereogram of a plagioclase crystal (optical symmetry planes) showing the position of extinction traverses (made with A_2) across the optical symmetry planes (figs. 3 and 4).

It should be noted that the biquartz becomes insensitive in the neighbourhood of an optic axis although extinction settings can be made.

Readings. With the objective iris open the crystal is brought to the centre of the field; the iris is closed and a symmetry plane is roughly adjusted. A_4 is locked at, say, 20° and the sensitivity of A_1 and A_2 is observed by making small rotations about those axes. If one of them produces a very rapid variation in the biquartz this setting of A_4 is reserved for adjusting that axis. If, on the other hand, this direction proves insensitive for A_1 or A_2 it is reserved for the other axis. A_4 is then reset to, say, 340° and a similar test is made; this normally yields a different relative sensitivity for A_1 and A_2. Thus it is possible to allot the adjustment of A_1 to one direction of A_4 and that of A_2 to the other, each being specially sensitive for the respective axis. The best condition is when one of the positions is completely insensitive, in which case the final adjustment by alternate settings can be very rapid.

When the biquartz has been matched at both the A_4 settings, the continuity of extinction is checked along the whole range of A_4, 330°–30°, and the readings for A_1 and A_2 are recorded.

Application. It is possible to use this procedure with ordinary extinction settings, but the biquartz is both more sensitive and more convenient, since it can be examined while the stage is stationary at the intended setting, whereas the position of maximum extinction can only be identified by making short alternate swinging movements about the axis under adjustment.

When the path-difference is large enough to give one or more complete rings in the interference figure the isogyre becomes comparable in width with the conoscopic reading mark and the conoscopic method, if practicable, will be preferred.

In many minerals the dispersion of the indicatrix may reach one degree; it is then essential to make the final adjustments with a monochromatic colour screen.

As a test for speed and accuracy, a series of planes in pairs at right angles were adjusted in various grains of a feldspar rock section. The average time for one plane was 2 minutes and the two great circles were rarely inclined at more than one degree from 90. Two of the grains presented all three symmetry planes; for these the values of $\Sigma \sin^2 A_2$ (Hallimond, 1950) were 1·0076 and 0·9911 for the hemisphere with n 1·54, and the three poles in each crystal fell within two degrees of the intersection of the great circles. The second of these crystals is shown in fig. 2. Figs. 3 and 4 illustrate the extreme changes in sensitivity for different inclinations about A_4. The extinctions plotted were measured on A_5, but the sensitivity for rotation of A_1 would not be very different. In fig. 2 the non-axial plane contains a position of insensitivity for A_2 near the middle of the arc (traverse at 20°, fig. 3); this unexpected result is in accordance with the Fresnel construction and is of great importance for the accurate adjustment of the feldspars. To adjust this symmetry plane in a plagioclase it will therefore be very useful to adjust A_1 with A_4 set at the middle of the arc YZ or YX, and A_2 at a position of A_4 nearer to an optical symmetry axis, i.e. nearer to the corner of the triangle (e.g. at 40° in fig. 2).

In fig. 4 the steep slope of those curves ($A_4 = 0°$ and 330°) that are nearest the optic axis reflects the narrowness of the isogyre at this position and the high sensitivity for A_2. At the axis the biquartz becomes insensitive and on passing through the axis the slope of the curves is reversed. Here it is best to adjust A_2 at a setting near the axis, say at

$A_4 = 0°$, while A_1 is adjusted with A_4 at a position nearer to the optical symmetry axis, say at 20° or 30°.

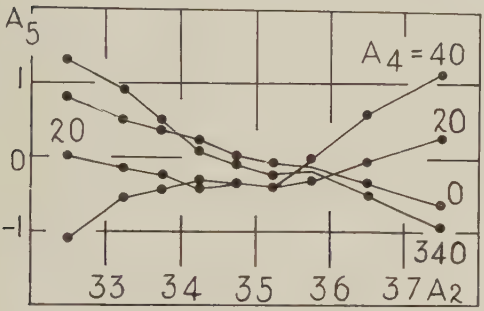


Fig. 3.

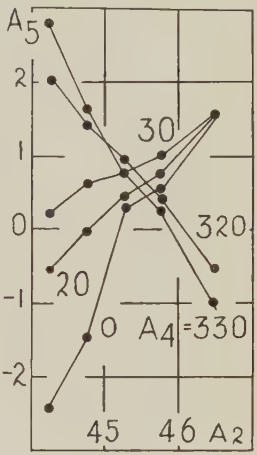


Fig. 4.

Fig. 3. Traverses across a non-axial symmetry plane in plagioclase (fig. 2). Extinction measurements on A_5 for various inclinations of A_2. The lines of traverse are marked in Fig. 2 with the readings of A_4 at which they were made.

Fig. 4. Traverses made as in fig. 3, across the axial plane in plagioclase (fig. 2).

Use of extinction curves. By special methods (e.g. N. Joel and I. D. Muir, 1958) it is possible to refine the location of a symmetry plane, but it may well be that the average of several settings made as here described would give a similar degree of accuracy. Even when extinctions are measured on A_5 the accuracy still depends upon the precise reading and the sensitivity of A_1 and A_2.

Joel and Muir plot the poles of the vibration directions, but it may be more convenient to plot extinction curves as in figs. 3 and 4. The curves as projected on the flat are in any case somewhat distorted, but since the only point to be determined is their intersection with a certain extinction value (that for the symmetry plane) the exact form of the rest of the curve is not material.

The axis A_3 used by Joel and Muir seems unnecessary, since it is equally easy to use A_2 as here described. Thus in fig. 3 the curves indicate a value for A_5 only slightly below $A_5 = 0°$; $A_2 = 35°$; this implies a negligible correction in A_1. In fig. 4 the indicated value for A_5 is $+\frac{1}{2}°$, $A_2 = 45\frac{1}{2}°$. A second set of curves showed that after an alteration of one degree in A_1 the intersection lay at $-\frac{1}{2}°$, with $A_2 = 45°$. It

should be noted that complete intersection is only obtained when the optical symmetry plane is within about one degree of correct adjustment. It may be doubted, however, whether these methods should be used, since it is less laborious to take the mean of several adjustments of the symmetry plane made with the biquartz as here described.

In the preceding account it has been assumed that the reader has already a normal acquaintance with the use of the stage, but details of the microscope adjustment are given because they differ very widely from those in some textbooks. The measurements were made on a Cooke, Troughton, and Simms three-axis Fedorov stage, and 'Research' microscope stand (A. F. Hallimond and E. W. Taylor, 1946 and 1950), and with a seven-degree biquartz, which was kindly prepared for the purpose by that firm.

Note added in proof. The existence of variations in sensitivity was discussed by F. J. Turner (1942) who proposed a new method for adjusting an optical symmetry plane (O.S.P.). As described by P. F. Kerr (1959) [summarized with Berek notation]: A_1 is rotated to extinction; A_4 is set to a position of maximum illumination; A_2 is set to minimum illumination; A_4 is returned to zero. Extinction should then be complete, indicating that an O.S.P. is in adjustment. Obviously this method will not meet all cases, but it often gives very rapid adjustment for plagioclase. The explanation appears to be that the crystal must have 2V approx. 90°. Then the Fresnel construction shows that the extinction direction of a section is nearly always within a few degrees of the azimuth of an O.S.P., which will thus lie approx. N–S after the initial rotation of A_1: the O.S.P. is then made vertical by rotating A_2 at a sensitive setting of A_4. The method described in the present paper relates to precise adjustment in the general case.

References.

Emmons (R. C.), 1931. In Winchell (A. N.), Microscopic characters . . . artificial minerals. John Wiley, New York.

Hallimond (A. F.), 1950. Mining Mag., vol. 83, pp. 12 and 77 [M.A. **11**–281].

—— 1953. Manual of the polarizing microscope; 2nd edn (York: Cooke, Troughton, and Simms).

—— and Taylor (E. W.), 1946. Min. Mag., vol. 27, p. 175.

—— —— 1950. Ibid., vol. 29, p. 150.

Joel (N.) and Muir (I. D.), 1958. Min. Mag., vol. 31, p. 860.

Kerr (P. F.). 1959. Optical mineralogy, 3rd edn, p. 119.

Turner (F. J.), 1942. Amer. Journ. Sci., vol. 240, p. 573.

—— 1947. Amer. Min., vol. 32, p. 390.

SHORT COMMUNICATION

Expanding-lattice minerals from Shropshire.

Unusual forms of expanding-lattice minerals have been found in a soil overlying the Cwm-mawr picrite in the Shelve area of south Shropshire, and their characteristics appear sufficiently distinctive to merit this preliminary note. The minerals occur in a brown earth near Woodgate, about ½ mile south of Corndon Hill.

In the (B)/C horizon of the profile pale yellowish-brown fibres and lath-shaped grains up to 4 mm. long resembling fragments of driftwood are fairly common: their form suggests that they are probably pseudomorphous after a fibrous mineral. In the partially weathered picrite underlying the profile there are cross-fibre veinlets of a mineral that when unweathered is green, but in the altered state is brown. The optical properties of the weathered fibres are closely akin to those of the soil mineral, and there seems no doubt that the latter represents the vein material in a more advanced state of alteration. Such veins appear to occur commonly and 'at Cwm-mawr, three-quarters of a mile west-north-west of Hyssington, weathered picrite traversed by half-inch veins of serpentine is seen' (Blyth, 1944). An examination of unweathered vein material from Woodgate and from Cwm-mawr shows that it is a fibrous chlorite with $a = 5{\cdot}3$ Å. as the fibre axis. The sharp basal reflections of 14·4 Å. periodicity are unaffected by heating below 600° C. or by treatment with glycerol, but heating at 700° C. shows changes in the relative intensities of the lower-order basal reflections characteristic of chlorites. The chlorite fibres have straight extinction and positive elongation, the refractive index being $1{\cdot}589 \pm 0{\cdot}003$ for light vibrating parallel to the fibre direction, and the fibre birefringence is approximately 0·004.

Microscopic examination shows that the soil mineral occurs in fibro-lamellar aggregates, slightly pleochroic in shades of yellowish brown, with the maximum absorption parallel to the length of the fibres. The refractive indices are variable but the maximum index is in the range 1·56–1·57 and the fibre birefringence is between 0·015 and 0·02. The zero, first, and second layers of the X-ray fibre pattern of air-dry material, separated from the soil after dispersion with Calgon (sodium hexametaphosphate), can be indexed on the basis of a cell with $a = 5{\cdot}3$ Å. (fibre axis), $b = 9{\cdot}2$ Å., $c = 15{\cdot}0$ Å., $\beta = 96$–$98°$. Reflections of

type $00l$, $0k0$, $0kl$, and hkl have been indexed. The occurrence of hkl reflections and reflections with $k \neq 3n$ shows that the layers are stacked in an ordered way. The long spacing of the fibres, 15 Å. in the air-dry state, is reduced to 10 Å. by heating at 110° C. and re-expanded to 15 Å. on rehydration. Treatment with glycerol alters the basal spacing to 18 Å. Throughout these changes a good fibre pattern is maintained, the gross features being unchanged although the details are altered.

The cation exchange capacity of 96 m.e. per 100 g. air-dry material has been obtained after saturation with calcium nitrate and replacement with magnesium nitrate solutions.

Another material in the weathering rock and the soil occurs as pale yellowish-brown waxy compound aggregates, but some grains show optical continuity and well-defined extinction directions. Many of the grains are lamellar owing to a well-developed cleavage and grains can be selected which give an excellent centred biaxial interference figure with $2V_\alpha$ 15–30°. The lamellar grains have straight extinction and pleochroism is slight with β and γ yellowish brown and α pale yellowish brown; the refractive indices are slightly variable but β is near 1·58 and the birefringence 0·02. Single-crystal rotation photographs about the normal to the lamellar faces show broad reciprocal lattice rods parallel to the rotation axis, which can be indexed on the basis of a hexagonal net with $a = 5{\cdot}3$ Å. or alternatively an orthogonal net with $a = 5{\cdot}3$ Å., $b = 9{\cdot}2$ Å. Oscillation photographs about an axis in the plane of the lamellar faces show a hexagonal array of diffuse spots (approximately the end projection of the hk rods) and a series of $00l$ reflections of a basal spacing of 15 Å. for air-dry material and 18 Å. for glycerol-treated material. There is also a considerable amount of less well-oriented material in each grain as the hk spots in each hexagon are joined by broad rings that follow constant θ lines. The X-ray data for the well-oriented material in these grains are consistent with a disordered stacking sequence of layers, the only regularity in addition to the equidistant stacking being the parallel alignment of the hexagonal a-axes of the half-layers of the mineral. On the basis of an orthorhombic structural unit, the well-developed cleavage is (001); the optic axial plane is (010) and the relationships between the crystallographic axes and the optical indicatrix are: $a = \gamma$, $b = \beta$, $c = \alpha$.

Caillière and Hénin (1951) have demonstrated the fibrous character and swelling properties of minerals classified as saponite, bowlingite, and diabantite. Fibre photographs taken here of saponite (bowlingite) from Cathkin Hills, Scotland, show an overall resemblance to those of

the fibrous soil mineral but there are differences in detail. The lamellar soil mineral seems very similar to the layer-lattice silicate found to be a constituent of iddingsite (Brown and Stephen, 1959), but differs in that the soil mineral can be readily expanded by glycerol.

It is likely that the fibrous mineral and the lamellar mineral owe their crystallographic order to the inheritance of undisplaced structural units from their parent minerals and that the study of such pseudomorphs will give structural information about clay minerals that could not be obtained from X-ray powder photographs or electron diffraction.

Pedology Department, G. Brown
Rothamsted Experimental Station, I. Stephen
Harpenden.

Blyth (F. G. H.), 1944. Quart. Journ. Geol. Soc. London, vol. 99 (for 1943), p. 187.
Brown (G.) and Stephen (I.), 1959. Amer. Min., vol. 44, p. 251.
Caillière (S.) and Hénin (S.), 1951. Clay Min. Bull., vol. 1, p. 138.

BOOK REVIEWS

Hawkes (H. E.). *Principles of geochemical prospecting.* Bull. U.S. Geol. Survey, 1957, no. 1000-F, pp. 225–355. Price 40 *c.*

Geochemical prospecting for minerals includes any method of mineral exploration based on systematic measurement of the chemical properties of a naturally occurring material. The purpose of the measurements is the location of geochemical anomalies or of areas where the chemical pattern indicates the presence of ore in the vicinity. Anomalies may be formed either at depth by igneous and metamorphic processes or at the earth's surface by agents of weathering, erosion, and surficial transportation.

Geochemical anomalies of deep-seated origin -primary anomalies—may result from (1) apparent local variation in the original composition of the earth's crust, defining a distinctive 'geochemical province' especially favourable for the occurrence of ore, (2) impregnation of rocks by mineralizing fluids related to ore formation, and (3) dispersion of volatile elements transported in gaseous form.

Anomalies of surficial origin—secondary anomalies—take the form either of residual materials from weathering of rocks and ores in place or of material dispersed from the ore deposit by gravity, moving water,

or glacial ice. The mobility of an element, or tendency for it to migrate in the surficial environment, determines the characteristics of the geochemical anomalies it can form. Water is the principal transporting agency for the products of weathering. Mobility is, therefore, closely related to the tendency of an element to be stable in water-soluble form. The chemical factors affecting the mobility of elements include hydrogen-ion concentration, solubility of salts, coprecipitation, sorption, oxidation potential, and the formation of complexes and colloidal solutions. The mobility of the elements may be further modified by biological factors.

Secondary anomalies may occur in residual materials or in materials transported by ice, frost, underground water, animals, soil-forming processes, plant activity, and surface water. Each one of these transporting agencies gives a characteristic distribution pattern to the weathering products of ore deposits.

Geochemical methods have been applied most extensively in the Soviet Union, Scandinavia, the United States, Canada, Africa, and Japan. The most uniformly successful geochemical prospecting work has been based on sampling and analysis of residual soil and vegetation; anomalies caused by movement of metals in ground and surface water show promise as an effective means of locating buried ore deposits. Some suggestions for the execution of geochemical surveys and the interpretation of geochemical data in terms of possible ore are presented.

KERR (Paul F.). *Optical Mineralogy.* London (McGraw-Hill Book Co., Inc.), 3rd edn, xiv+442 pp. Price 66*s*.

Part I (Optics) is the same length as in the 2nd edition and the chapter headings have been retained, but the text has been largely rewritten. Among microscopes the Zeiss Standard is described, but the new large Leitz models are not represented. A new chapter has been inserted, on the Universal stage (13 pp.). Conoscopic procedure and the three-axis stages are not mentioned. The four-axis stage is described with an Emmons notation, but the five-axis stage is shown with a notation which uses the well-known Berek symbols in reversed order (see Emmons, Amer. Min., 1929, **14,** 441). Since this method was expressly discarded by Emmons (Geol. Soc. Amer., 1943, 8, 13) as 'renumbering the axes, leading to tragic confusion' its revival is unfortunate; the Berek, Reinhard, and Emmons notations are established and may well suffice. A few details may need correction: p. 5; epoxy resins (A. G. King, Amer. Min., **42,** 689, 1957) are important for impreg-

nating sections. Fig. 2.7 is much less clear than the usual diagrams of the microscope. Fig. 2.20: this mica compensator does not tilt.

Reflected light methods and polishing have been purposely omitted, and there are very few references to artificial products. The book thus retains the petrological trend of the earlier editions and will be chiefly useful to petrologists working with transparent thin sections.

Part II forms an excellent introduction to thin-section petrology. The crisp but small micrographs of the 2nd edition have been replaced by numerous good photographs, with many optical diagrams. Clay minerals are described, with some electron micrographs. References at the end of each chapter include many textbooks and monographs. The somewhat high price reflects the good quality of the production. The inclusion of a general first part follows usual practice, but it may be questioned whether these varied subjects might not now be allotted to separate textbooks.

A. F. H.

Rose (A. J.). *Tables et Abaques.* Paris (Centre National de la Récherche Scientifique), 1957, 141 pp. Price 1500 francs.

Durif-Varambon (A). & Forrat (F.). *Tables numériques de sin θ et de $sin^2\theta$.* Centre National de la Récherche Scientifique, Institut Fourier, Grenoble, France, 1958, 26 pp. Price 250 francs.

Rose's main table (III) lists d-values for X-ray powder photographs for Bragg angles, θ, between 0° and 90° at intervals of 0·01°. Values are given for the $K\alpha_1$ wavelengths of the usual radiations (from Cu, Ni, Co, Fe, Cr, and Mo targets). The wavelengths taken (table I) are those of Cauchois and Hulubei, converted to absolute Ångströms; they differ from the usual values by not more than 1 part in 90 000, which is insignificant for most purposes. The tables are thus directly comparable, for example, with those of the National Bureau of Standards (1950, Applied Mathematics Series, 10); random checks show that they agree to the limit above.

A new and valuable feature of this volume is that table III also lists values of $4R\theta$ and $2R\tan\theta$ for each value of θ; $R = 180/2\pi$ or $240/2\pi$ applying to standard American or French cameras of 5·73 cm. or 7·64 cm. diameter respectively. $4R\theta$ represents the distance between powder lines on a cylindrical camera, and can be applied to measurements on a focusing camera; $2R\tan\theta$ gives the corresponding distance measured on a flat-film camera and can be applied to measurements of

layer-line repeat distances on rotation photographs taken in either the normal or the Mauguin mode. These quantities can obviously be applied also to cameras of diameter 11·46 cm. or 15·28 cm. diameter by simple proportion; neither quantity gives rounded values at 0·01° intervals of θ, but the intervals are sufficiently short to make interpolation easy.

Both these and the Nat. Bur. Stand. tables list d-values for $K\alpha_1$ wavelengths only, and are hence not strictly applicable to normal powder photographs, for which $K\alpha_m$ wavelengths are required at low Bragg angles. In the present case this is because these tables are intended primarily for users of curved quartz-crystal monochromators (with focusing cameras). They include a useful nomogram relating the radius of curvature of the crystal with its distances from the target-focus and the camera-focus for each of the above wavelengths and for various angles of cut between the crystal surface and the $(10\bar{1}1)$ plane of the quartz crystal.

Table II*a* lists atomic weights and mass absorption coefficients for the elements, while table II*b* gives values of e^{-x} to four decimals in the ranges $x = 0(0{\cdot}01)5{\cdot}50$ and $5{\cdot}50(0{\cdot}10)9{\cdot}90$. The format and legibility of all the tables are excellent.

The tables of Durif-Varambon and Forrat are for users of X-ray powder cameras 'de 240 mm de périmètre' (not 'de diamètre' as stated on the cover). They list θ, $\sin\theta$, and $\sin^2\theta$ for values of $4R\theta$ (see the review above) in the range 20(0·1)240 mm. ($R = 38{\cdot}2$ mm.). Sin θ and $\sin^2\theta$ are given to five decimal places; θ occurs at intervals of $2\frac{1}{4}$ minutes of arc on the above scale. The tables are thus designed for a very narrow public, since this interval of θ is awkward for interpolation by users of other cameras. There is indeed a need for five- (or six-)figure tables of $\sin^2\theta$. It seems a pity that the authors did not adopt the format of Rose's tables, or better that their values of $\sin\theta$ and $\sin^2\theta$ were not incorporated in them. Perhaps this would be possible in a future edition.

R. J. Davis

Borchert (H.). *Ozeane Salzlagerstätten*. Berlin (Gebrüder Borntraeger). 1959, viii+237 pp., 27 text-figs. Price DM. 48.

This book is an excellent successor to Professor Borchert's earlier works on salt deposits. It presents a masterly review of the complex history of marine evaporites, from their initial deposition to their later reconstruction by geothermal metasomatism, leaching by groundwaters, and other processes.

The first part of the book is devoted to depositional factors. A

discussion of the general climatic, topographic, and tectonic conditions, with reference especially to the Zechstein evaporites, is followed by sections on the separation of salts from sea water under static isothermal conditions, and under 'dynamic polythermal' conditions where temperatures vary in different parts of the depositional area. These involve the calcium salts and the relevant part of the system Na–K–Mg–Cl–SO_4–H_2O. Metastable phenomena are considered.

The following 73 pages deal with post-consolidation metasomatic changes, including the various stages of metasomatism of important salt parageneses by solutions, resulting from melting with rise of temperature and pressure on burial, migrating from upper layers to lower ones and vice-versa, and by solutions produced by groundwater leaching or from juvenile waters. The evidence for metasomatism in salt deposits is discussed in detail, and includes the occurrence of mineral phases and parageneses unstable under conditions of primary deposition, textural evidence (pseudomorphs, &c.), veins and impregnations, blue halite, and certain types of facies change. The processes are discussed in relation to experimental work, and the principal changes in the Zechstein evaporites are given.

The rest of the book includes comparisons of oceanic and non-oceanic deposits; sections dealing with the evaporites of the Rhine graben and some other deposits; the association of salt with oil; and a discussion of salt tectonics and the mechanical properties of salts. There is a large and useful bibliography (40 pp.).

The book would be improved by the addition of more text-figures (for example, maps showing facies changes in the Zechstein), but this is the only slight qualification in my admiration for a work which contains a very large amount of information, and deals with a highly complex subject in a way that makes it essential to all workers interested in evaporites and in metasomatic processes.

F. H. Stewart

Konta (J.). *Jílové minerály Československa.* [*Clay Minerals of Czechoslovakia.*] Česk. Akad. Věd, Prague, 1957, 319 pp., 165 figs., 65 tables. Price 35·50 Kčs.

This excellent volume gives details of the clay mineralogy of some twenty-two clay occurrences in Czechoslovakia. After a general description of clay minerals, their structure, classification, and nomenclature, and the methods of investigation used, the remainder of the

book is divided up on the basis of the mineralogy of the deposits studied. Thus, three occurrences of allophane, sixteen of kandites, eight of smectites, eight of clay micas (including dillnite), and three of sepiolite and palygorskite are dealt with. The data given on each sample is remarkably exhaustive, consisting of macroscopic and microscopic data, differential thermal and dehydration curves, X-ray diffraction data, electron micrographs, and in most instances chemical analyses. The description of each sample examined is completed by some remarks on its genesis. An extensive bibliography and very adequate name and subject indexes are given. The book is excellently produced, and electron micrographs and X-ray powder photographs show detail adequately, despite the fact that art paper is not used. The binding is, however, hardly up to the standard of the remainder of the book, and it is doubtful if it would stand up to much handling.

Dr. Konta is to be congratulated for his energy and for the tremendous amount of work which he must have undertaken to produce such an excellent survey, and one can only feel disappointed that no comparable book is available for other countries: it is, in a more limited field, comparable with Heddle's classical and monumental 'Mineralogy of Scotland'. Despite the fact that the book is written in Czech, and that workers in other countries may not have great interest in the actual deposits described, there is such a wealth of detail on individual minerals that all clay mineralogists will find much to interest them.

R. C. Mackenzie

Swineford (A.), editor. *Clays and Clay Minerals.* Proceedings of the Fifth National Conference on clays and clay minerals, University of Illinois, 1956. Washington (National Academy of Sciences—National Research Council, Publication 566), 1958, vii+360 pp., 156 text-figs. and pls., 58 tables. Price $4.50.

The National Clay Committee of the National Academy of Sciences (U.S.A.) was formed some seven years ago and has since then held seven annual meetings at various centres in the U.S.A., publishing the papers presented at these meetings in a yearly volume entitled 'Clays and Clay Minerals'; the one under review is the fifth of the series. Since the National Clay Committee has deliberately attempted to interest not only clay mineralogists, chemists, and physicists, but also all interested in clay from any angle, practical or theoretical, the twenty-eight papers included in this book cover a very wide range and defy a simple classification into sections. Nevertheless, they are all of great interest to clay

mineralogists. Thus, in one paper it is suggested that halloysite is not tubular in nature but that the tubes are artifacts formed on drying, in another that pure montmorillonite may contain two distinct entities differing in charge, and in another that the clay minerals found in deposits depict the source conditions rather than the deposition conditions. Such physical subjects as the surface charge on montmorillonite and the density of montmorillonite pastes, as well as such practical aspects as the stratigraphical associations of clay minerals and the alteration of clay minerals in sea water, are also treated. The volume clearly reflects the vast amount and the wide scope of current American investigations on clays. Each paper is provided with a concise summary that truly indicates the scope of the paper and the conclusions reached —a very refreshing change from the type of summary so often presented. The electron micrographs reproduced are in general excellent, although a few seem to be hardly up to the previous high standard of this series. One misses any account of discussion on the various papers, but the general quality of the book more than makes up for this deficiency; a brief but adequate index is provided. The volume is excellently produced and is, indeed, a pleasure to handle. The Editor is to be congratulated on the high standard obtained, and the Committee on providing a book no clay mineralogist can afford to be without.

R. C. Mackenzie

Kraus (Edward Henry), Hunt (Walter Fred), & Ransdell (Lewis Stephen). *Mineralogy. An Introduction to the Study of Minerals and Crystals.* London (McGraw-Hill Book Co., Inc.), 5th edn, 1959, x+686 pp. Price 70*s*.

The general arrangement, chapter headings, and much of the subject matter and illustrations of this very well produced text-book are the same as in the third edition [1936; M.A. **6**–289], but the text has been reset with many minor alterations and some of the less satisfactory figures have been dropped. The book well deserves its continued popularity.

M. H. H.

Hintze (Carl) [1851–1916]. *Handbuch der Mineralogie.* Chudoba (Karl F.). *Ergänzungsband II: Neue Mineralien und neue Mineralnamen (mit Nachträgen, Richtigstellungen und Ergänzungen).* Berlin (Walter de Gruyter & Co.), 1955–9, Lief. 6–9, pp. 401–728. Price, Lief. 6 & 7 DM. 22 each, Lief. 8 DM. 28, Lief. 9 DM. 26.

In the sixth part the alphabetical review of new minerals, new mineral

names, and new data is completed (tungomelan to zirkonoid; cf. M.A. 12–564). A very useful critical and classified arrangement of all the new names in Erg. I and II follows; accepted, doubtful, synonymous, and other names are carefully distinguished, though Chudoba's classification in this respect will not be universally acceptable. The seventh and eighth parts contain a review of rejected and doubtful minerals, suggested and proven identities, arranged alphabetically; this very useful compilation, which is styled 'Ergänzungsband II, Teil II', serves admirably to show how many problems of identity and nomenclature are still unsettled. Finally, part 9 is the first part (Abernathyit to Iochroitt) of 'Ergänzungsband II, Teil III', which will really be a third appendix in itself, containing new species and varieties, new names, rejected and doubtful names, suggested and proven identities, and new data, for the period 1954–8 inclusive, arranged alphabetically. It is unfortunate that so many names have been given only in a Germanized version, without cross-indexing under the original spelling; this has led to one of the few slips detected in the work: Kryophiolith (p. 572) and Cryphiolith (p. 691) are the same mineral, crifiolate of Scacchi.

M. H. H.

Brauns (R.) [1861–1937]. *Mineralogie.* Chudoba (Karl F.). *Allgemeine Mineralogie.* Sammlung Göschen, Bd. 29. Berlin (Walter de Gruyter & Co.), 1958, 10th edn, 120 pp., 120 figs. Price DM. 3.60. *Spezielle Mineralogie.* Sammlung Göschen, Bd. 31/31a. Idem, 1959, 10th edn, 170 pp., 125 figs. Price DM. 5.80.

The first edition of this little text-book appeared as a single volume in 1893, and the 7th in 1936 [M.A. 6–339]; the 9th edition (1955) and the present edition have been greatly expanded and divided into two volumes, one giving an elementary general account of crystal growth, form, and structure, of the simpler methods for the qualitative analysis of minerals, and of the conditions of formation of minerals; the second volume comprises concise descriptions of the commoner species. There is no account of crystal optics (which is the subject of a separate volume in the Sammlung Göschen), and optical data for the species described are not given under the individual species, but collected in one table. The pair of booklets (6 in. × 4 in.) are clearly written in simple language and are excellent value.

M. H. H.